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KONINKLIJKE AKADEMIE
VAN WETENSCHAPPEN
-:- TE AMSTERDAM -:-

PROCEEDINGS OF THE

SECTION OF SCIENCES

VOLUME XII

MO. BOT. GARDEN
1910

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JOHANNES MÜLLER :—: AMSTERDAM
:═══════: JULY 1910 :═══════:

(Translated from: Verslagen van de Gewone Vergaderingen der Wis- en Natuurkundige Afdeeling van 29 Mei 1909 tot 29 April 1910. Dl. XVIII.)

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VAN WETENSCHAPPEN
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VOLUME XII
(— 1ST PART —)

JOHANNES MÜLLER :—: AMSTERDAM
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KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Saturday May 29, 1909.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van Zaterdag 29 Mei 1909, Dl. XVIII).

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Geophysics. — *"On the determination of tidal constants from observations performed with horizontal pendulums."* By Dr. J. P. VAN DER STOK.

(Communicated in the meeting of April 23, 1909).

1. When applying the theory worked out with astronomical accuracy of the analysis of tidal observations we are checked in practice by disturbing influences of meteorological nature which may be great with respect to the quantities to be calculated, making thus the accuracy of the theory as well as that of the results illusory.

These disturbing influences are greatest when we calculate the tides of long duration, where meteorological factors of irregular nature play such a preponderant part, that the definition of the constants is only then possible if it is based upon series of such long duration that those disturbances can be regarded as eliminated. Each investigation ought then to be preceded by a study of those disturbances and a taxation based upon the latter of the demand to be put as a minimum for the length of the series.

For most tides of short duration these objections do not hold, or at all events in a much less degree, they do however for those tides whose period differs but little from that of mean solar time and which are connected with it in a systematic way, namely the sidereal tides proper K_1 and K_2 and the sidereal tide improper P.

As the disturbances of meteorological nature appearing here have not only an irregular but also a regular character, the former can be eliminated by making the calculation over a great many years, but not the latter and only by means of more or less uncertain hypotheses shall we be able to come closer to our aim.

Thus e. g. the tides K_1 and P can only then be calculated out of the observations if we assume that the diurnal meteorological tide S_1 remains constant during the whole year, a supposition which is certainly inaccurate because the mean amplitudes of the motion S_1, finding its origin in the land- and seawind, must be considerably less in winter than in summer.

As lucky circumstance can be regarded that S_1 is generally small a. o. on European coasts, where however on the other hand K_1 and P too, are abnormally small, abnormally, namely, with respect to the value evaluated out of the theory of equilibrium. In tropical regions S_1 is in many places not small, and only where, as in the Java sea, K_1 and P are great, an approximative determination of K_1 is possible,

whilst for that of the small tide P the circumstances will be unfavourable always and everywhere.

2. In a recent publication [1]) Prof. HECKER has made an investigation of the influence of forces generating tides on two horizontal pendulums set up for this aim in a pit so that the influence of changes in temperature was reduced to a minimum.

The result of this investigation surpasses the expectations about the possibility of such a determination of disturbances of gravity caused by celestial bodies which were until a short time ago decidedly unfavourable.

The two instruments are not of equal value; the pendulum set up in an azimuth of 222° proves to be a much better instrument than the other; the results obtained with the best pendulum are, expressed in deviations of an equivalent vertical pendulum in arc seconds

M_2	Calculated	$0''.00922\ cos\ (2\ t - 305°.5)$
	Observed	$0''.00622\ cos\ (2\ t - 285°.4)$
S_2	Calculated	$0''.00399\ cos\ (2\ t - 305°.5)$
	Observed	$0''.00244\ cos\ (2\ t - 273°.6)$

Deviations in the sense of the movement of the hand of a timepiece are reckoned positive for this instrument.

HECKER concludes from this that the earth deforms itself under the influence of the forces of the tides and that its rigidity or resistance against deformation is about equal to that of a steel sphere. This result is in accordance with the conclusions drawn from the variation of the velocity of propagation of the first forerunners of a seismic wave (longitudinal vibration) from 7 to 13 K. M. and of the second forerunners (transversal vibration) from 4 to 7 K. M. with the depth of the layers passed through and likewise with the fact that the principal periodical oscillation of the geographical latitude does not take place in a period of 306 d. (EULER) but of 427 d. (CHANDLER), from which would ensue a still greater resistance against deformation, namely about twice that of steel.

In the last chapter HECKER discusses in short the possibility to deduce from the data the influence of the partial tidal force K_1, diurnal periodical with sidereal time; here however he is checked by the above mentioned difficulty that evidently there exists in the diurnal periodic movement S_1, (Table I) not of astronomical origin, an annual variation.

---

[1]) O. HECKER. Beobachtungen an Horizontalpendeln über die Deformation des Erdkörpers unter dem Einfluss von Sonne und Mond. Veröffentl. des Kön. Preuss. Geod. Inst. Neue Folge, no. 32, 1907.

He therefore regards an arrangement according to sidereal time not recompensatory and from a proof taken by shifting of the monthly means he concludes:

"Immerhin können wir aber aus den angeführten Werten entnehmen, dass die Amplitude einer etwa vorhandenen Sternzeitwelle nur sehr gering sein kann".

TABLE I.

Diurnal inequality in the movement of the pendulum No 1.

Commencement of time: Midday. M. E. time.

January	$0''.0021$	$\cos (t - 270^\circ.0) +$	$0''.0029$	$\cos (2t - 321^\circ.9$
February	65	249.4	43	307.4
March	97	274.7	52	300.0
April	142	260.7	35	281.6
May	153	254.6	20	249.8
June	196	242.0	7	164.0
July	190	241.4	15	165.1
August	188	246.5	19	244.8
September	208	234.0	42	250.7
October	165	233.4	45	259.7
November	137	231.5	29	262.1
December	34	259.7	25	319.3
Year	$0''.01296 \cos (t - 245''.4) +$		$0''.00244 \cos (2t - 277^\circ.5)$	

In consequence of this discussion we can remark: 1. that for an accurate calculation of the sidereal tides an arrangement according to sidereal time, proper or improper, is unnecessary, so that little trouble is attached to it, and 2 that the influence of those sidereal tides cannot be so insignificant.

Besides the sidereal-time solar-lunar tide proper K_1 also the solar tide P appears which moves with respect to the mean solar time in the same way as sidereal time but in an opposite sense.

The angular velocity of K_1 is:

$15^\circ + 0^\circ.04107$ an hour

that of P:

$15^\circ - 0^\circ.04107$ „ „

The theoretic coefficient of importance [1]) expressed in the mean amplitude of M_2 as unity is

for K_1: 0.58
for P: 0.19

so that yearly twice the two tides strengthen each other, namely in June and in December, when the relative amplitude is: 0.77, whilst in March and in September it comes to: 0.39.

There must therefore be, even though the meteorological tide S_1 were constant during the whole year, a considerable annual variation in the diurnal inequality.

Although during the period under treatment the lunar declination was particularly small and the circumstances therefore were unfavourable for the determination of the constants of the sidereal tides, yet the theoretical amplitude of K_1, expressed in deviations of the vertical pendulum, amounts for Potsdam to:

$$0''.0050$$

whilst the amplitude of P is in square numbers:

$$0''.0020$$

The amplitude of the annual variation in the diurnal inequality must therefore be about:

$$0''.0070$$

i. e. almost twice that in the principal solar tide S_2:

$$0''.0040.$$

From the amplitudes of the diurnal movement of Table I is evident that the amplitude of the annual variation:

$$\text{June—December} = \frac{196-34}{2} = 0''.0081$$

differs but little from the theoretical value, so that we have every reason to make an attempt for an accurate determination of the constants, which promises to lead, at all events for the tide K_1, to satisfactory results.

The value of such an investigation is not so much to be found in the determination itself as in the fact that, if the investigation is continued over several years so that the irregularities of meteorological origin have disappeared, we shall be able to correct the monthly means of the diurnal inequality for the influence of the astronomical tides, in order to obtain in this way accurate series of numbers from which the nature and the origin of the S_1 tide can be studied and deduced.

---

[1]) Sir George Darwin. Scientific papers, Cambridge. 1907, vol. I. p 25.

3. The amplitude of a vertical pendulum under the influence of a celestial body can be represented by the expression

$$H\,p\,q\,sin\,2\,\zeta,$$

ζ = distance of zenith,

H = general coefficient or greatest amplitude which the celestial body could effect on a place situated under the equator with a declination zero:

Expressed in arc seconds H is for the moon $= \frac{3}{2} \times \frac{206265''}{2\,fk^3} = 0.''01737,$

H' for the sun $= 0.4604\,H,$

if we put:

$f = 81.4$ = quotient of mass of earth by mass of moon.

$k = 60.26$ = mean distance from moon to earth, in earthradii.

p = geographical or local coefficient, dependent on the geogr. latitude of the place of observation and the azimuth of the horizontal pendulum; for the rest different for diurnal and semidiurnal movements.

q = astronomical coefficient dependent on the inclination of the orbit and its eccentricity.

If the general expression is developed in a series of terms, behaving itself purely periodically, we find for the components in the direction of the meridian, N (north positive), and in a direction normal to it, W (west positive), as far as the terms are concerned, in the direction of sidereal time, inclusive of P, for the moon:

$$\left.\begin{aligned} N &= H\,q_m\,cos\,2\,\varphi\,sin\,\gamma\,\tau \\ W &= -\,H\,q_m\,sin\,\varphi\,cos\,\gamma\,\tau \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad . \quad (1)$$

φ = geogr. latitude

$\gamma = 15° + \eta = 15°.04107$ an hour

$q_m = sin\,I\,cos\,I$ (I = inclination of orbit of moon)

and for the sun:

$$\left.\begin{aligned} N &= H'\,cos\,2\,\varphi\,[q_s\,sin\,\gamma\,\tau - q'\,sin\,(\gamma - 2\,\eta)\,\tau] \\ W &= H'\,sin\,\varphi\,[-\,q_s\,cos\,\gamma\,\tau + q'\,cos\,(\gamma - 2\,\eta)\,\tau] \end{aligned}\right\} \quad . \quad . \quad (2)$$

The deviation of a horizontal pendulum set up in an azimuth $180° + \alpha$ is:

$$-\,(N\,sin\,\alpha + W\,cos\,\alpha)$$

If we put:

$$-\,sin\,\varphi\,cos\,\alpha = p\,sin\,\chi$$
$$-\,cos\,2\,\varphi\,sin\,\alpha = p\,cos\,\chi,$$

then for Potsdam:

$$\varphi = 52°\,23', \qquad \alpha = 42°$$
$$p = 0.6129, \qquad \chi = 286°.15, \qquad q' = 0.4127.$$

For the period treated (Dec. 1902—April 1905) holds:

$$I = 18^\circ\,33', \qquad v = 0^\circ.31$$

with which we find with the aid of the wellknown tables of BÖRGEN for the astron. coefficient of the K_1 tide composed of (1) and (2)

$$q'' = 0.4732,$$

from which follows for the theoretical value

$$K_1 = 0''.005037, \qquad P = 0''.001869$$

4. Already 15 years ago I have pointed out in "Studiën over Getijden in den Indischen Archipel", that for the determination of the constants of the tides K_1, P and K_2 the trouble of an arrangement of the hourvalues according to the angular velocities of these tides is superfluous and that we can deduce these constants with equal accuracy and little labour directly out of the monthly means. The application of this method has furnished good results not only for the Indian tides but also for the determination of the tidal constants on the Dutch coasts where all three tides are very small.[1]) Especially if as in this case hour-observations are at hand, the calculation is exceedingly simple, for the diurnal variation can be represented in its variability in the course of the year with great approximation by the expression:

$$S_1 \cos(15t - C_1) + K_1 \cos(15t - \psi_1 + 30x) + P\cos(15t - \psi_2 - 30x), \quad . \quad (3)$$

where

$$\psi_1 = \chi_1 - V_0 \quad , \quad V_0 = h_0 - v' - \frac{\pi}{2} = 204^\circ.71 \, ,$$

$$\psi_2 = \chi_2 - V'_0 \quad , \quad V'_0 = -h_0 + \frac{\pi}{2} = 155^\circ\,16 \, ;$$

h_0 = length of the sun at the commencement of the time (epoch) (294°.84) i. e. in this case January 16; v' is a small correction, caused by the inclination of the orbit of the moon with respect to the ecliptic. It is clear that the inaccuracies committed here, namely 1 the angular velocities holding for *one* day being all taken equal instead of respectively

$$15^\circ \quad , \quad 15^\circ + \eta \quad , \quad 15^\circ - \eta$$

And 2 the monthly means being regarded as 12 equidistant points, can have no perceptible influence on the result of the calculation.

Omitting the first term we can bring (3) into the form:

---

[1]) Études des phénomènes de marée sur les côtes néerlandaises I. Utrecht, 1904.

$$cos\ 15\,t \left[\begin{array}{r} (K_1\ cos\ \psi_1 + P\ cos\ \psi_2)\ cos\ 30\ x \\ +(K_1\ sin\ \psi_1 - P\ sin\ \psi_2)\ sin\ 30\ x \end{array} \right]$$

$$+\ sin\ 15\,t \left[\begin{array}{r} (K_1\ sin\ \psi_1 + P\ sin\ \psi_2)\ cos\ 30\ x \\ -(K_1\ cos\ \psi_1 - P\ cos\ \psi_2)\ sin\ 30\ x \end{array} \right]$$

If we put the formula for the diurnal inequality of Table I into the form :

$$A\ cos\ 15\ t + B\ sin\ 15\ t,$$

and subtract from the values A and B the annual means A' and B' and then again represent the differences formed in this way by :

$$\left. \begin{array}{l} A - A' = R\ cos\ 30\ x + Q\ sin\ 30\ x \\ B - B' = R'\ cos\ 30\ x + Q'\ sin\ 30\ x \end{array} \right\} \quad . \quad . \quad . \quad . \quad , \quad . \quad (4)$$

we obtain the equations :

$$\left. \begin{array}{r} K_1\ cos\ \psi_1 + P\ cos\ \psi_2 = R \\ K_1\ sin\ \psi_1 - P\ sin\ \psi_2 = Q \\ K_1\ sin\ \psi_1 + P\ sin\ \psi_2 = R' \\ -K_1\ cos\ \psi_1 + P\ cos\ \psi_2 = Q' \end{array} \right\} \quad . \quad . \quad . \quad . \quad . \quad , \quad . \quad (5)$$

from which the four unknown quantities can be solved ; then the amplitudes must be augmented, monthly means having been used, by multiplication by the factor :

$$R_2 = \frac{\pi}{12\ sin\ 15^\circ} = 1.0115$$

and to the values ψ_1 and ψ_2 the astronomical arguments V_0 and V'_0 must be added.

In all cases in which the S_1 tide is so small that even if it is submitted to an annual variation it can only exercise an influence small with respect to the amplitudes of K_1 and P, this simple method leads to good results.

If S_1 is not small, we can start from the assumption that land- and seawind are different in winter and summer, but that they can be regarded as constant during each of the seasons. We can then eliminate out of the six summer months of the differences (4) the value $S_1\ cos\ (15\,t - C_1)$ and likewise out of the six winter-differences, and then we can calculate out of the equations the four unknown quantities. The combinations of the different monthly means necessary for this are of course less favourable than in the former case, but this disadvantage can be compensated by taking a great number of years together which is necessary for every method when we have to deal with disturbances of a meteorological nature.

5. Neither of the two methods, which can be used when we calculate watertides, is applicable to the case under discussion; as evidently the tide S_1 is great we may not assume that the annual variation of S_1 will be small with respect to the quantities to be found. Neither is there a reason to assume that in winter or in summer, regarded separately, S_1 will be fairly constant, as we are still quite uncertain about the nature and the origin of it.

Another favourable circumstance however, not appearing for watertides, makes in this case an approximative solution possible.

For when K_1 appears in the monthly means with a certain value K and an argument χ_1, then we can say almost with certainty that the tide P will make its appearance with an amplitude:

$$P = aK,$$

in which a is the theoretic proportion of P to K_1:

$$a = 0.371.$$

Furthermore we can state with equal probability

$$\chi_2 = \chi_1 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

Thus out of the equations (4) two unknown quantities disappear and they can be replaced by two others characterizing S_1 more closely.

We represent this tide by the form:

$$\{S + L\cos(30\,x - m)\}\cos(15\,t - C_1)$$

and so we assume that the amplitude is submitted to an annual variation, but that C_1 remains constant, which, if this phenomenon finds its origin in the radiation of the sun, cannot be far from the truth.

Furthermore follows from (6):

$$\psi_2 = \psi_1 + V_0 - V'_0 = \psi_1 + \alpha = \psi_1 + 49°.55.$$

Instead of the formulae (5) we get the four equations

$$\left.\begin{aligned}
L\cos C\cos m + K\cos\psi_1 + Ka\cos(\psi_1+\alpha) &= R\\
L\cos C\sin m + K\sin\psi_1 - Ka\sin(\psi_1+\alpha) &= Q\\
L\sin C\cos m + K\sin\psi_1 + Ka\sin(\psi_1+\alpha) &= R'\\
L\sin C\sin m - K\cos\psi_1 + Ka\cos(\psi_1+\alpha) &= Q'
\end{aligned}\right\} \quad . \quad . \quad (7)$$

With

$$\begin{aligned} R &= 42.22 & R' &= 69.94\\ Q &= 34.98 & Q' &= 0.96\\ C &= 245°.5 \end{aligned}$$

we find from this:

$$\begin{aligned} K_1 &= 51.2 & L &= 46.3\\ \psi_1 &= 44°.5 & m &= 248°.7 \end{aligned}$$

or, after applying the augmenting factor R_2 and the astronomical argument, as the result of this investigation:

$$K_1 \begin{cases} \text{Calculated} & 0''.00504\ cos\ (t - 286°.2) \\ \text{Observed} & 0''.00518\ cos\ (t - 249°.2) \end{cases}$$

$$P \begin{cases} \text{Calculated} & 0''.00187\ cos\ (t - 286°.2) \\ \text{Observed} & 0''.00192\ cos\ (t - 249°.2) \end{cases}$$

and then for the monthly means of the diurnal inequality the expression

$$0''.01296\ \{1 + 0.361\ cos\ (30\ x - 248°.7)\}\ cos\ (15\ t - 245°.5)$$

6. Just as for watertides the superposition of the partial tides S_2 and K_2 causes a maximum in March and September (the equinoctial tides known to every mariner) and a minimum in January and July, here too such a semi-annual variation must appear in the expressions for the semi-diurnal inequality. Indeed this variation makes its appearance clearly at first sight in the amplitudes of Table I.

If we assume that the S_2 tide is constant during the whole year, then the general expression for the monthly means is:

$$S_2\ cos\ (30\ t - C_2) + K_2\ cos\ (30\ t - C_{2k} + 60x)$$

where

$$C_{2k} = \chi - V_0 \quad , \quad V_0 = 2\ h_0 - 2\ v''.$$

We have then to do nothing but to analyse the expressions of Table I, after subtraction of the mean for the whole year, into the components:

$$A\ cos\ 30\ t + B\ sin\ 30\ t$$
$$A = cos\ (60\ x - C_{2k})$$
$$B = -\ sin\ (60\ x - C_{2k})$$

If we bring through the differences A and B doubly-periodic curves, we obtain for each of the quantities to be found:

$$K_2\ sin\ C_{2k} \text{ and } K_2\ cos\ C_{2k} \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

two values which must be about equal and from which we deduce, after applying the augmenting factor

$$R_4 = \frac{\pi}{6\ sin\ 30°} = 1.0472$$

and the astronomical argument

$$V_0 = 2\ h_0 - 2\ v'' = 229°.68 - 0°.25,$$

the quantities K_2 and χ.

The astronomical coefficient calculated according to the tables of BÖRGEN is:

$$q = 0.0878$$

and if we put

$$-\sin\varphi\cos\alpha = p\sin\chi$$
$$\sin\varphi\cos\varphi\sin\alpha = p\cos\chi$$

we then find for the theoretical value:

$$K_2 = 0''.00085 \quad , \quad \chi = 305°.5.$$

From the data of Table I we find, however, for the values (8) quantities, which are equal in sign but which differ for the rest pretty much, namely:

$$K_2\cos C_{2k} = \begin{cases} 3.97 = R \\ 10.77 = Q \end{cases}$$

$$K_2\sin C_{2k} = \begin{cases} 4.10 = R' \\ 16.34 = Q' \end{cases}$$

So here too we must assume that S_2 is not constant and that there is a semi-annual variation in the semi-diurnal inequality which indeed is immediately evident from the amplitudes of Table I on account of the inequality of the two maxima and minima.

If, as above, we represent the S_2 tide by

$$\{S_2 + L\cos(60\,x - m)\}\cos(30\,t - C)$$

we then find out of the four equations

$$L\cos C\cos m + K_2\cos C_{2k} = R$$
$$L\cos C\sin m + K_2\sin C_{2k} = Q$$
$$L\sin C\cos m + K_2\sin C_{2k} = R'$$
$$L\sin C\sin m - K_2\cos C_{2k} = Q'$$

as the result of the investigation:

$$K_2 \begin{cases} \text{Calculated} & 0''.00085\cos(2\,t - 305°.5) \\ \text{Observed} & 0''.00070\cos(2\,t - 260°.6) \end{cases}$$

and for the semi-diurnal inequality:

$$0''.00244\{1 + 0.574\cos(60\,x - 158°.6)\}\cos(2\,t - 277°.7)$$

This result justifies the expectation that if monthly means of the diurnal variation are available calculated over a greater number of years and by preference over years in which the declination of the moon is great, also the calculation of the small tide K_2 can be made with all the looked for accuracy. For, at the greatest declination of the moon the amplitude of K_2 is almost twice greater than at the smallest.

Out of the monthly means of the diurnal variation for the second horizontal pendulum set up at Potsdam these sidereal tides do not admit of a deduction. The continual displacements of the zero point, considerable for both instruments, surpass for this instrument all the small regular movements entirely.

A determination of the siderial tides might thus serve as a criterion for the quality of seismic instruments.

Mathematics. — "*The constructive determination of the velocities of a spacial system*". By Prof. J. CARDINAAL.

(Communicated in the Meeting of April 23, 1909).

1. In Vol. LVI page 315, 1908 of the Zeitschrift für Mathematik und Physik Prof. K. TH. VAHLEN inserted an article on the graphic composition of forces in space, where he introduces the observation, that where such forces must be composed, we have so far occupied ourselves with general geometric considerations and have set aside the possibility of executing them. He points to the fact, that it is important to place oneself on the standpoint of descriptive geometry and to arrange the operation in such a way that the construction has to be made in two perpendicular planes of projection. At the same time he develops a method for forces in space and gives an example of it. The same observation can be made for an other part of mechanics, namely, for the theory of motion. In this paper we shall treat of the constructive solution of a problem out of the theory of motion in space.

2. It is known that the motion of an invariable spacial system is determined as soon as we know that five points of the system must move over five indicated surfaces. The normals of the surfaces in these points are five director lines of a focal system, whose principal axis is at the same time the axis of the motion. If moreover we know the length of the velocity of one of these points, then the velocities of all the others are known in length and direction. It requires a great number of lines to execute this construction; we shall therefore somewhat simplify the form of the problem and give to it the following wording:

Given are three fixed points A, B, C of a spacial system; A moves along the right line a, B along the line b; C must remain on a definite plane. Of point A is still given the velocity AA_v in a definite position. Assuming this position we wish to construct:

a. The axis of the movement.

b. The velocities of the points B and C.

3. Passing on to the execution (Fig. 1) we assume the plane in which C moves to be the horizontal plane of projection; we then bring through a the plane normal to the preceding one and we choose

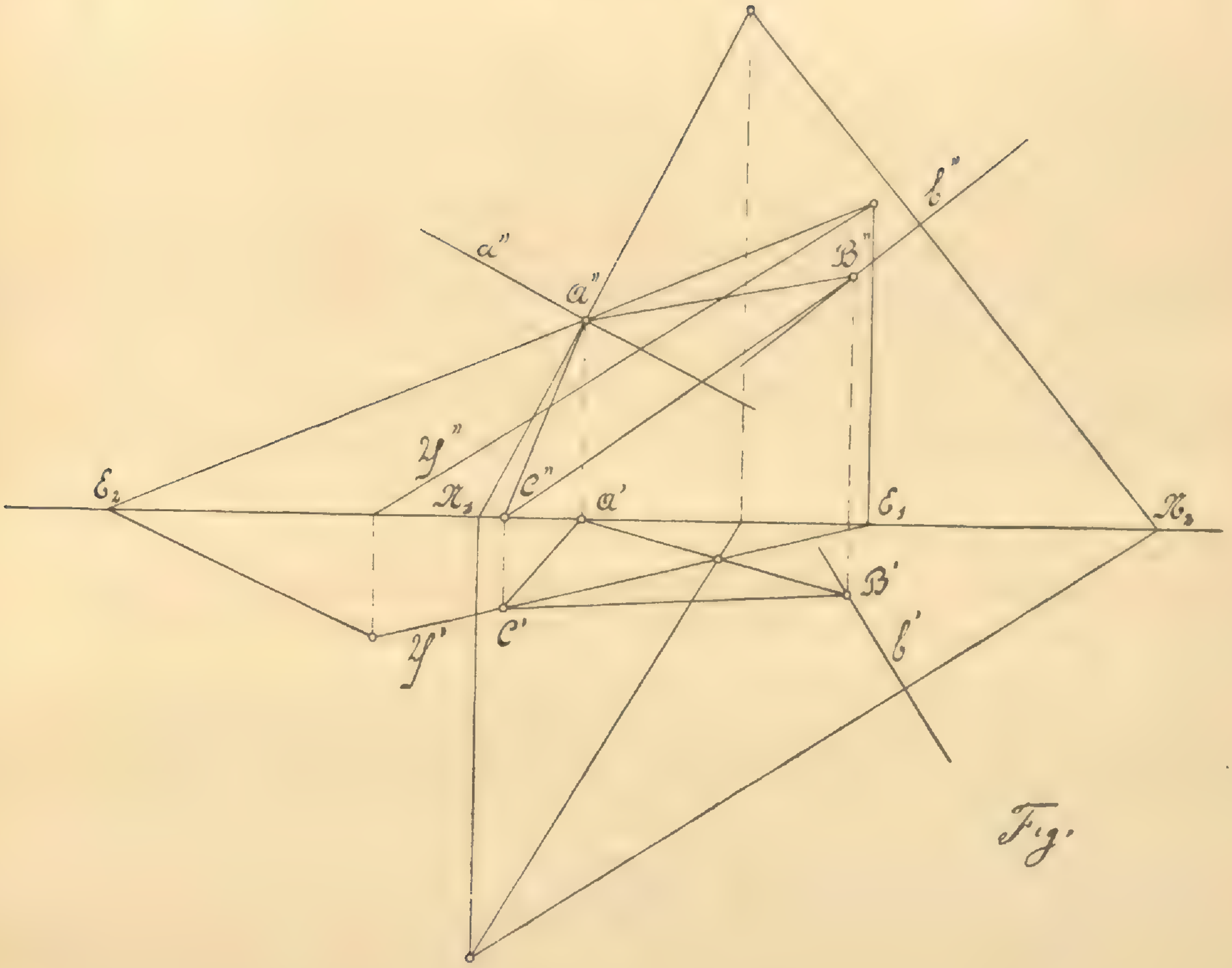

Fig.

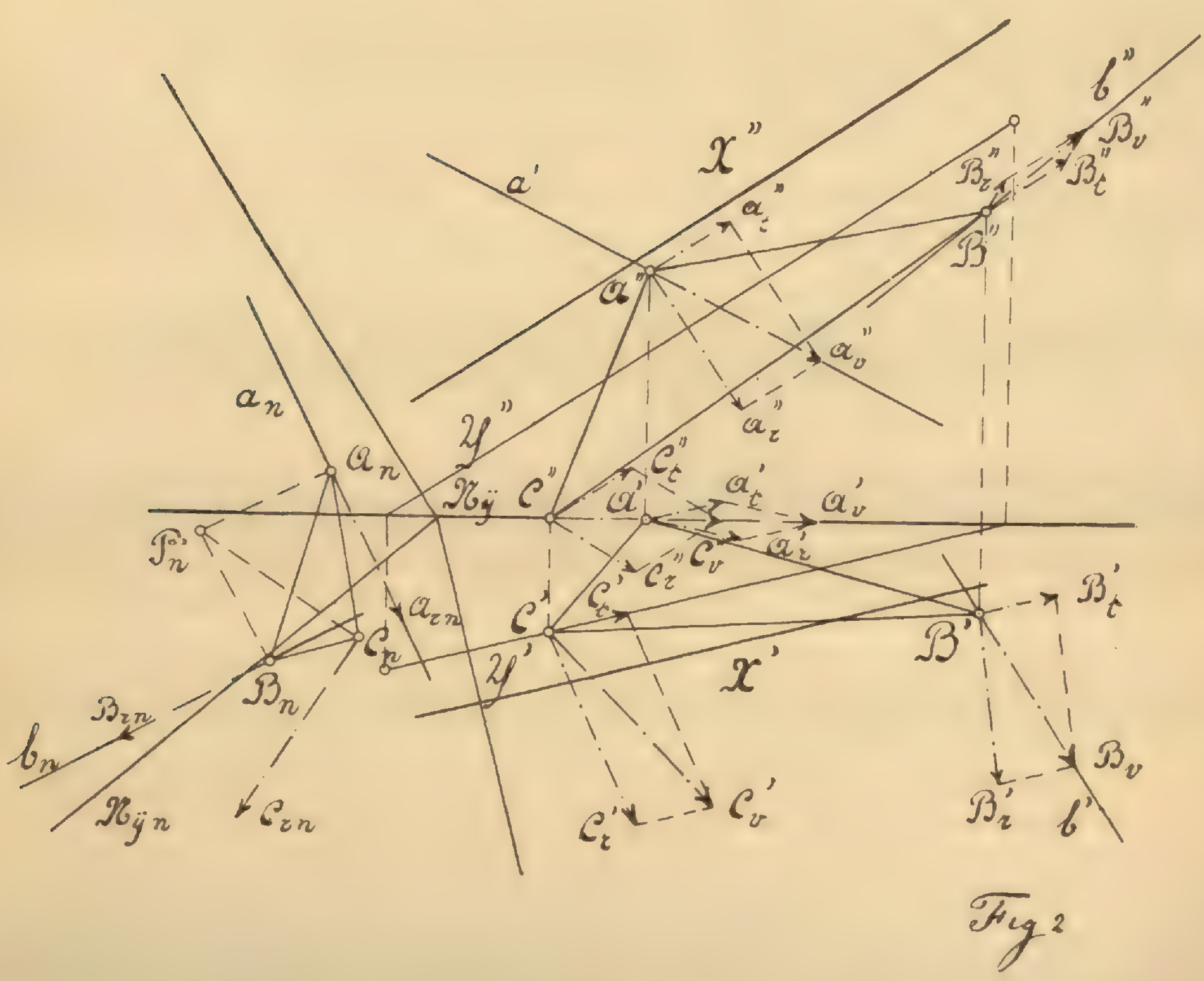

Fig 2

this as vertical plane of projection; the position b has now been indicated by two arbitrary projections. Through A we bring the plane $N_1 \perp a$, through B the plane $N_2 \perp b$; the line of intersection $(N_1 N_2)$ is conjugate to AB. Let us construct the transversal of $(N_1 N_2)$ and AB normal to the horizontal plane and let us bring the plane E_1 through C and that transversal. The line of intersection Y of the plane E_1 with the plane E_2, made parallel to AB and $(N_1 N_2)$ runs parallel to the principal axis of the focal system, at the same time axis of the motion.

4. We shall deduce out of Y the real position of the principal axis and to that end we choose fig. 2 [1]), upon which $\triangle ABC$ together with the line of intersection have been transferred. We put a plane $N_y \perp Y$ and we project on it the points A, B, C with the directions of velocity a and b of A and B. In the figure these are represented revolved into the vertical plane by A_n, B_n, C_n, a_n, b_n. Let us erect out of A_n, B_n the normals on a_n, b_n; these intersect each other in the pole P_n of the motion projected on N_y. The pole P_n is also a revolved point of the principal axis; from this follow easily the projections X', X'' of these.

On N_y we now also project A_v; in the figure $A_n A_{rn}$ is the revolved projection of AA_v; as the pole P_n is known we can construct the revolved projections of BB_v and CC_v on N_y; from this follows length and direction of the components BB_r and CC_r in the planes normal to the axis X; the components $// X$, AA_t, BB_t, CC_t which must be equal to each other, are now also easy to find.

5. Although when arranging this construction we have only spoken of the lines a and b and the horizontal plane we can generalize the supposition by assuming that a and b are the tangents of the points A and B of the curves described by two fixed points of the spacial system and that the horizontal plane is the tangential plane to C of a curved surface described by a third fixed point.

Farthermore may be observed that when only the velocity of point B needs constructing it is sufficient to construct the direction Y, the real position of the axis X not being necessary.

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[1]) The two figures separated from each other are derived from a single figure executed on a large scale by Mr. G. FERGUSON, assistant at the Technical University at Delft.

Chemistry. — *"Javanese Basilicum oil and methylchavicol"*. By Prof. P. VAN ROMBURGH.

(Communicated in the meeting of April 23, 1909).

In December 1900 I had the honour to communicate to the Academy the results of a preliminary research, which I had carried out in conjunction with Dr. TROMP DE HAAS, as to the composition of the essential oil of a variety of *Ocimum Basilicum* known as *selasih hidjau*.

It was shown that this oil consists mainly of methylchavicol.

Since then, I have procured larger quantities of this oil, and was thus enabled to isolate a few more components, and to find out more in particular whether an olefinic terpene may be present, which I had succeeded in isolating in large quantities from a related species (or variety).

The oil investigated showed $D_{14} = 0.962$ and $\alpha_D = + 1°20'$ ($l = 2$ d.m.). From 1500 cc., about 210 cc. were separated by distillation in water vapour. These were fractionated in vacuo.

Up to 88° were obtained 75 cc. ($\alpha = 9°17'$ $l = 2$ d.m.)
from 88—93° „ „ 105 „ ($\alpha = 1°16'$ $l = 2$ d.m.)
residue 26 „ .

The distillate up to 88° was once more fractionated and 28 cc. were collected boiling below 70° ($D = 0.856$) and 63 cc. boiling at 70°—85°.

As the fraction with the lowest b.p. had an odour of cineol it was freed from this by shaking with a 50% solution of resorcinol[1]). The fraction boiling at 70°—85° was also treated in the same manner. The layer floating on the resorcinol, amounting to 21 cc., was again fractionated in vacuo and finally, a fraction was obtained boiling at 60°—70°, $D_{15} = 0.8208$. The quantity, however, was too small to admit of a further separation. The density and also the odour of this fraction render it very probable that it contains indeed an olefinic terpene, probably ocimene, mixed with a cyclic one.

From the resorcinol solution, which had served for the elimination of the cineol, the latter was readily isolated by distillation in steam. The density ($D_{18} = 0.926$) and the reaction with iodol proved the identity. BERTRAM and WALBAUM had already shown the presence of cineol in German Basilicum oil and also in the Basilicum oil from Réunion [2]).

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[1]) Semi-Annual Report of SCHIMMEL & Co., Oct. 1907. p. 32.
[2]) Arch d. Pharm. **235**, 176 [1897].

The methylchavicol, which was obtained by fractional distillation from Javanese Basilicum oil, still showed a faint rotation (1°30′ in a 2 d.m. tube).

In order to eliminate the active ingredient, which could not be separated by fractional distillation, it was treated with various reagents such as acetic anhydride, sodium hydrogen sulphite, potassium permanganate, but all in vain; the rotation did not diminish. A treatment with magnesinm-ethyl iodide, which was repeated a few times, finally answered the purpose. The boiling point of the main mass was found to be 214°—215° (corrected).

Analysis: found 79.88 C, 8.26 H; calculated 81.08 C, 8.1 H.

During the fractional distillation of methylchavicol it will be noticed that the residue in the flask often deposits some crystals.

These may be obtained in a somewhat larger quantity by heating methylchavicol in sealed tubes for 48 hours at 250°. The contents of the tubes has then turned yellow. After distillation up to 240° (thermometer in the liquid) the residue is placed in an ice chamber. A crystalline mass is formed from which the oil is removed by suction. After repeated recrystallisation from acetone and alcohol the main product isolated melts at 98° [1]), while a substance melting at 166° is also obtained. The solutions of these substances in petroleum ether and in benzene exhibit a beautiful blue fluorescence which is particularly strong in the case of the substance melting at 166°. Traces of a substance melting above 200° were also formed, but the yield of these crystallised substances is rather small.

The elementary analysis gives the same results with the two first named substances and agrees with the analysis of methylchavicol.

The determination of the molecular weight of the substance melting at 98° in acetone with Rᴜʙᴇʀ's apparatus gave 308, 311 and 308. A determination in chloroform gave 280. The substance melting at 166° gave, in chloroform, 293, 290 and 279 as molecular weight. As the molecular weight of methylchavicol is 148, these substances should therefore be dimerides of the same.

The substance melting at 98° combines with 1 mol. of bromine and forms a compound melting at 87°.

Analysis: found 35,04 Br; calculated 35.05 for $C_{20}H_{24}O_2.Br_2$.

The investigation of these polymerides, which up to the present is only of a preliminary nature, owing to the great difficulties connected with their preparation, is being continued.

Utrecht. Lab. org. Chem. University.

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[1]) The complete purification of this product is very troublesome and the melting point is not very sharp.

Chemistry. — *"The essential oil from the fruits of Morinda citrifolia L."* By Prof. P. VAN ROMBURGH.

(Communicated in the meeting of April 23, 1909).

Morinda citrifolia L, a plant from the family of the *Rubiaceae* (Malay : Bengkudu, Sundanese : Tjangkudu) occurs in the wild state in Java and is also cultivated for the sake of the red dye, which can be obtained from the root bark. The over ripe fruits of this shrub have a very disagreeable odour, reminding of rancid fat. Some years ago, when at Buitenzorg, I prepared from it an essential oil by distillation in water vapour, which was subjected in 1896 to a preliminary investigation by my then assistant C. J. E. LOHMANN [1]), which showed it to contain a higher volatile fatty acid, accompanied by an ester.

During my stay at Buitenzorg, last summer, I found back an old specimen of the oil and Dr. A. W. K. DE JONG had the great kindness to prepare for me the essential oil from 1000 Kilos of Bengkudu fruits, for which I wish to express my gratitude, so that I had sufficient material for an investigation.

A portion of the essential oil was separated by decantation from the aqueous distillate, but in other cases the oil was obtained by shaking the aqueous distillate with benzene.

The oil separated by decantation had a brownish-yellow colour and was turbid owing to minute, glittering, crystals floating therein. The specific gravity was 0.927 at 13°. The sp.gr. of another sample, of which only a small quantity was at my disposal, was 0.961 at 14°.

The crystals were filtered off and the filtered oil treated with a dilute sodium hydroxide solution, in which it is almost entirely soluble, while a turbid liquid is formed from which a thin oily layer separates after a very long time. Shaking with ether or benzene readily causes emulsions, which separate with difficulty. The least troublesome way to effect the separation of the volatile neutral components from the substances dissolved in the alkali was found to be a careful distillation of the dilute solution.

With the aqueous vapours a liquid, the odour of which is not disagreeable, passes over, which floats on water. The alkaline liquid, which was heated in a basin on the waterbath, in order to remove the last traces of volatile admixtures, was strongly acidified with sulphuric acid and a yellow layer of liquid acids collected on the surface. When these had been removed the liquid was distilled to

[1]) Report Gov. Botanical Gardens, Buitenzorg 1896 p. 59.

recover any dissolved acids. A few drops of slightly soluble fatty acids were recovered.

The separated acids, after having been dried over anhydrous sodium sulphate were subjected to a careful fractional distillation, which yielded two fractions boiling constant within a few degrees, one at about 205° and another at 238°. Of acids having a higher boiling point only traces are present.

The liquid boiling at 205° solidified when cooled in liquefied ammonia. The melting point was situated at — 5°.2, $D_{15} = 0.932$. These constants agree with those of n. capronic acid [1]).

The acid boiling at 238° solidified to a beautiful crystalline mass at a temperature below 15°. The melting point was 15°.2, $D_{14} = 0.913$. n. Caprylic acid [2]) has the same constants; addition of caprylic acid m.p. 15° from the specimen collection of the laboratory did not cause any melting point depression.

The amount of caprylic acid in the oil investigated was about eight times that of the capronic acid.

The content in neutral volatile constituents amounts to a few per cent only and consequently I had at my disposal such a small quantity that a separation by fractional distillation gave no sharp results. Fractions were obtained boiling at about 170°, 196° and 208°.

These were again united and heated with strong aqueous potassium hydroxide. After dilution with water a very slight quantity of an oily liquid separated which resisted a further treatment with the alkali. The alkaline liquid was heated in a flask to obtain the alcohols formed. The distillate gave, on addition of potassium carbonate, a separation of about 0.5 cM.[3] of a mixture of alcohols, which commenced to boil at about 70° and in which the presence of ethyl alcohol could be demonstrated with certainty (iodoform reaction) whilst the presence of methyl alcohol [3]) is very probable. Moreover, an odour of fusel oil is noticed in the fraction having the higher b.p.

The acid mixture formed in the saponification contains mainly caprylic acid, which was isolated in a pure condition, and to judge from the b.p. also a slight quantity of capronic acid.

The oil of the fruits from *Morinda citrifolia* therefore contains

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[1]) Georg W A. Kahlbaum, Zeitschr. phys. Ch. XIII, 40 [1894], gives the m.p. as — 5°.2, the b.p. as 205°.7 (corr.).

[2]) For n. caprylic acid Georg. W. A. Kahlbaum (loc. cit p. 62) gives the m.p. as 15 .1, the b.p. as 237°.5. Schey, Rec. **18**, 185 [1899] found the m.p. = 16°, and $D_{20} = 0.910$.

[3]) Methyl alcohol occurs in abundant quantity in the *durian* fruits (from *Durio zibethinus* Murr.).

ethyl caprylate and capronate in small quantities and to judge from the b.p. also a little of the methyl esters of the same acids.

The crystals separated from the oil were recrystallised from boiling alcohol in which they are but sparingly soluble. The m.p. is 60°.

The elementary analysis gave a result showing that this substance is a saturated hydrocarbon (found C 85.2 H 15.2).

Sulphuric acid has no action, even on being heated; its solution in chloroform does not decolorise a bromine solution, whilst potassium permanganate in acetone does not react.

Parraffins, m.p. 55°—65°, not unfrequently occur in essential oils, on the other hand n. capronic- and n. caprylic acid but rarely occur in the free state in such oils and then only in minute quantities. The essential oil from the fruits of *Morinda citrifolia* which consists of more than 90 % of these acids is, therefore, very remarkable in that respect.

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Mathematics. — "*On pentaspheric geometry.*" By Dr. S. L. VAN OSS. (Communicated by Prof. P. H. SCHOUTE.)

(Communicated in the meeting of April 23, 1909).

1. Referred to a rectangular simplex of coordinates $OX_1X_2X_3X_4$ and to an imaginary unity point whose cartesian coordinates are: $x_1 = x_2 = x_3 = x_4 = i$, the equation

$$x^2_1 + x^2_2 + x^2_3 + x^2_4 + x^2_5 = 0 \quad . \quad . \quad . \quad . \quad . \quad (1)$$

represents a real hypersphere H in fourdimensional space S_4.

Let X be a point of H, X' the stereographic projection of X on $x_4 = 0$, then, if x, y, z, be the non homogeneous coordinates of X' referred to the axes OX, OY, OZ, coinciding respectively with OX_1, OX_2, OX_3, we have the following relations:

$$x_1 : x_2 : x_3 : x_4 : x_5 = 2x : 2y : 2z : x^2 + y^2 + z^2 - 1 : \frac{x^2+y^2+z^2+1}{i}$$

Putting

$$2x = x'_1 \ , \ 2y = x'_2 \ , \ 2z = x'_3 \ , \ x^2+y^2+z^2-1 = x'_4 \ , \ x^2+y^2+z^2+1 = ix'_5,$$

we have identically

$$x'^2_1 + x'^2_2 + x'^2_3 + x'^2_4 + x'^2_5 = 0 \quad . \quad . \quad . \quad . \quad . \quad (1)$$

These x'_i are known to represent a set of pentaspheric coordinates referring X' to the five orthogonal spheres:

$$x = 0 \ , \ y = 0 \ , \ z = 0 \ , \ x^2 + y^2 + z^2 - 1 = 0 \ , \ x^2 + y^2 + z^2 + 1 = 0.$$

Omitting dashes we establish analytical identity between the geometry of the hypersphere H and that of pentaspheric space Π.

2. *Corollaries.*

I. *One* linear equation in pentaspheric point-coordinates:

$$\Sigma\, \xi_2\, x_3 = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

represents a *sphere* Ξ, this being the stereographic projection of the section with H of the S_3 that has the same equation.

The coefficients ξ_i are the coordinates of Ξ.

II. *Two* linear equations:

$$\Sigma\, \xi_i\, x_i = 0 \quad , \quad \Sigma\, \eta_i\, x_i = 0 \quad . \quad . \quad . \quad . \quad . \quad (3)$$

represent a circle Ξ/H.

III. *Three* equations:

$$\Sigma\, \xi_i\, x_i = 0 \quad , \quad \Sigma\, \eta_i\, x_i = 0 \quad , \quad \Sigma\, \zeta_i\, x_i = 0 \quad . \quad (4)$$

represent the *cowple of points* $\Xi/H/Z$.

IV. *Four* equations:

$$\Sigma\, \xi_i\, x_i = 0 \quad , \quad \Sigma\, \eta_i\, x_i = 0 \quad , \quad \Sigma\, \zeta_i\, x_i = 0 \quad , \quad \Sigma\, \theta_i\, x_i = 0, \quad . \quad (5)$$

have in general no geometrical equivalent.

V. Ξ is a *zero-sphere* if the corresponding S_3 is tangent to H, which is the case if

$$\Sigma\, \xi_i^2 = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

VI. Two spheres Ξ and H intersect orthogonally if the corresponding spaces S_3 are conjugate with respect to H, i.e. when

$$\Sigma\, \xi_i\, \eta_i = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

3. Now considering in their mutual connection the relations:

$\Sigma\, x_i = 0$: *identity* between pentaspheric point-coordinates, (1)

$\Sigma\, \xi_i\, x_i = 0$: *equation* of the sphere Ξ, ... (2)

$\Sigma\, \xi_i = 0$: condition that Ξ *zero-sphere*, ... (6)

$\Sigma\, \xi_i\, \eta_i = 0$: „ „ two spheres cut orthogonally, (7)

the following we may remark.

We may occasionally consider a set of *independent* x_i to represent a *sphere* X having the same coordinates as a point X in S_4; now this sphere is the stereographic projection of the section of the *polar space* of the point X with the fundamental hypersphere.

Viewing matters thus, the relation $\Sigma\, x^2 = 0$ appears as the *equation of a three-dimensional manyfold of zero-spheres*; the space Π appearing as a *four-dimensional space of spheres*.

At the same time $\Sigma\, \xi_i\, x_i = 0$ appears as the condition that the spheres Ξ and X intersect orthogonally.

Considered as *equation in x-variables*, this relation determines the sphere Ξ as the representative image of the complex of x-spheres cutting it orthogonally, if considered as *equation in* ξ*-variables*, it determines the sphere X as the representative image of a complex of ξ-spheres cutting it orthogonally.

Thus, pentaspheric analysis disengaging itself from the impeding identy (1), *the space* Π *rises to the rank of a linear four-dimensional space in which*, **in virtue of the established, now completed, correspondence with** S_4, *x-spheres and* ξ*-spheres appear as dual elements.*

4. *Table of Correspondence.*

In pentaspheric space Π.		*In* S_4.
x-Sphere-element		Point
Pencil	*of x-spheres*, to be represented by the *limiting points*	Line
Net	„ „ „ „ „ „ „ *orthogonal circle*	Plane
Complex	„ „ „ „ „ „ „ „ ξ*-sphere*	S_3
ξ*-Sphere-element*		S_3
Pencil	*of* ξ*-spheres* „ „ „ „ „ *common circle*	Plane
Net	„ „ „ „ „ „ „ „ *couple of points*	Line
Complex	„ „ „ „ „ „ „ *orthogonal x-sphere*	Point

In virtue of this choice of representative images, correspondence between S_3 and ξ-sphere, plane and circle, straight line and couple of points, such as has been established at first, remains unaltered, it has only been rendered complete, now that the x-sphere appears as the image of the arbitrary point in S_4.

The application of these principles reaching all over projective geometry of S_4, pentaspheric geometry in its generalised form may be characterized as the *projective geometry* of our space Π, which does not differ essentially from that in S_4.

With this is stated as a matter of fact, that with *special* exploration of the space Π it is all over now.

We have still to show how *construction* in Π is connected with that in S_4.

Some few simple examples we are going to deal with, may show that geometrical figures in Π corresponding to such in S_4, as admit effective construction with the aid of *ruler only*, may be determined by a set of constructions requiring only the ordinary use of *ruler and compasses.*

5. To this end we call the attention to some properties concerning

the STEFANOS *pentacycle* and the SEGRE *configuration*, two closely connected figures.

Referring to SEGRE's memoir in *Rendiconti di Palermo, Vol.* II, and to COSSERAT's in *Annales de Toulouse Vol.* III, we may assert that the STEFANOS *pentacycle* and SEGRE's *associated sets* are essentially equivalent figures. In fact, SEGRE shows that the 5 transversal planes to six straight lines form an associated set ; herewith is stated that the common elements to 6 complexes of planes in S_4 are the transversal planes to the axes of the degenerated complexes appearing in the linear system which is determined by 6 complexes.

(Hence also follows that these axes constitute the cubic variety with 10 double points, studied by SEGRE).

Hence we immediately deduce *that the pentacycle may be obtained by projecting stereographically the section of a set of associated planes with a hypersphere.*

But now also appears the dual match to the pentacycle: *a set of associated couples of points*, the same being the stereographic projection of the section of a set of associated lines with a hypersphere.

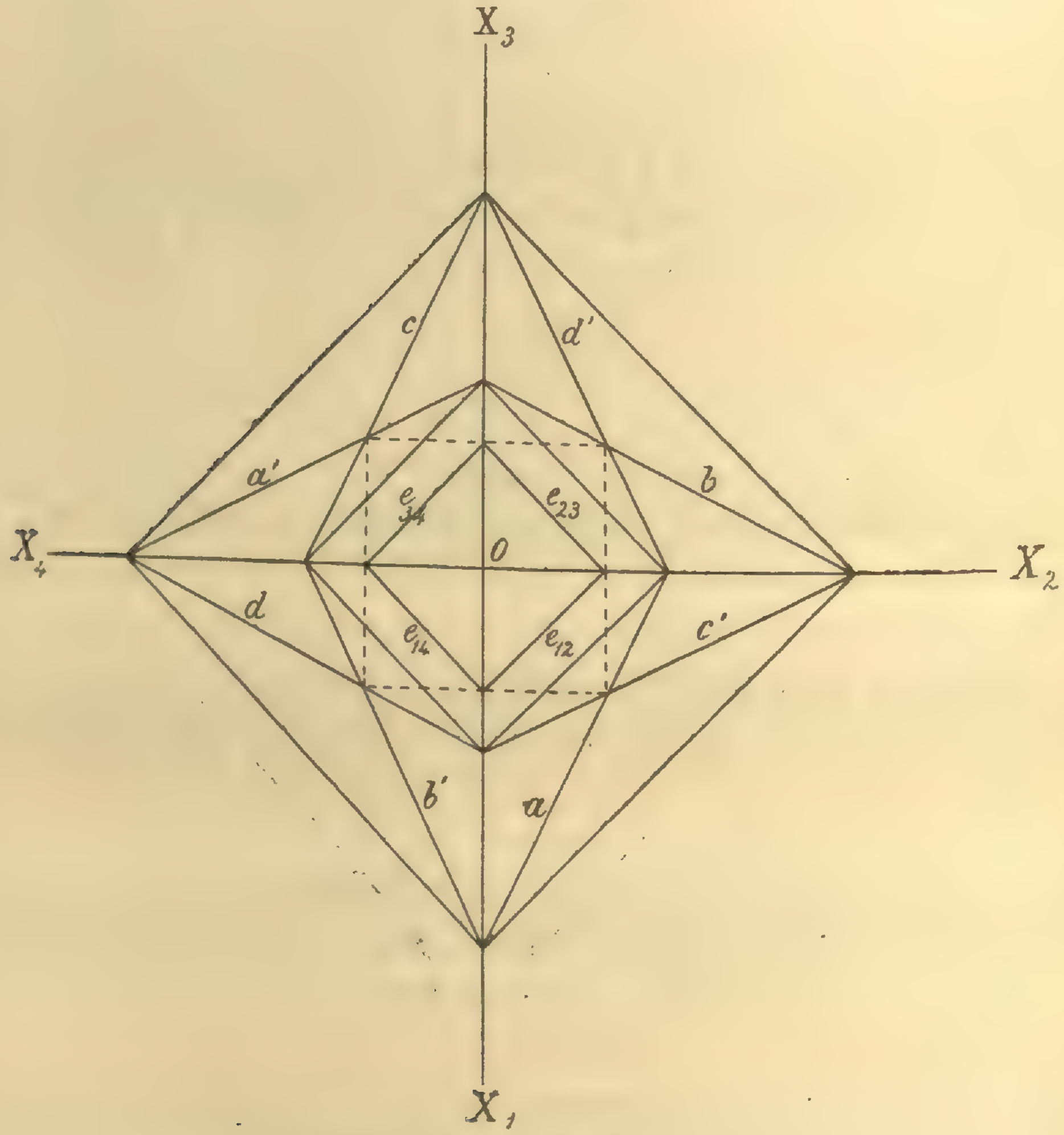

Fig. 1

In the same way the two dual forms of the SEGRE configuration get their image in our space.

That this correspondence really leads to a practical construction of the pentacycle or its dual figure will be apparent from the extreme simplicity of the figures 1 and 2 showing the SEGRE sets $a\,b\,c\,d\,e$, $\alpha\,\beta\,\gamma\,\delta\,\varepsilon$.

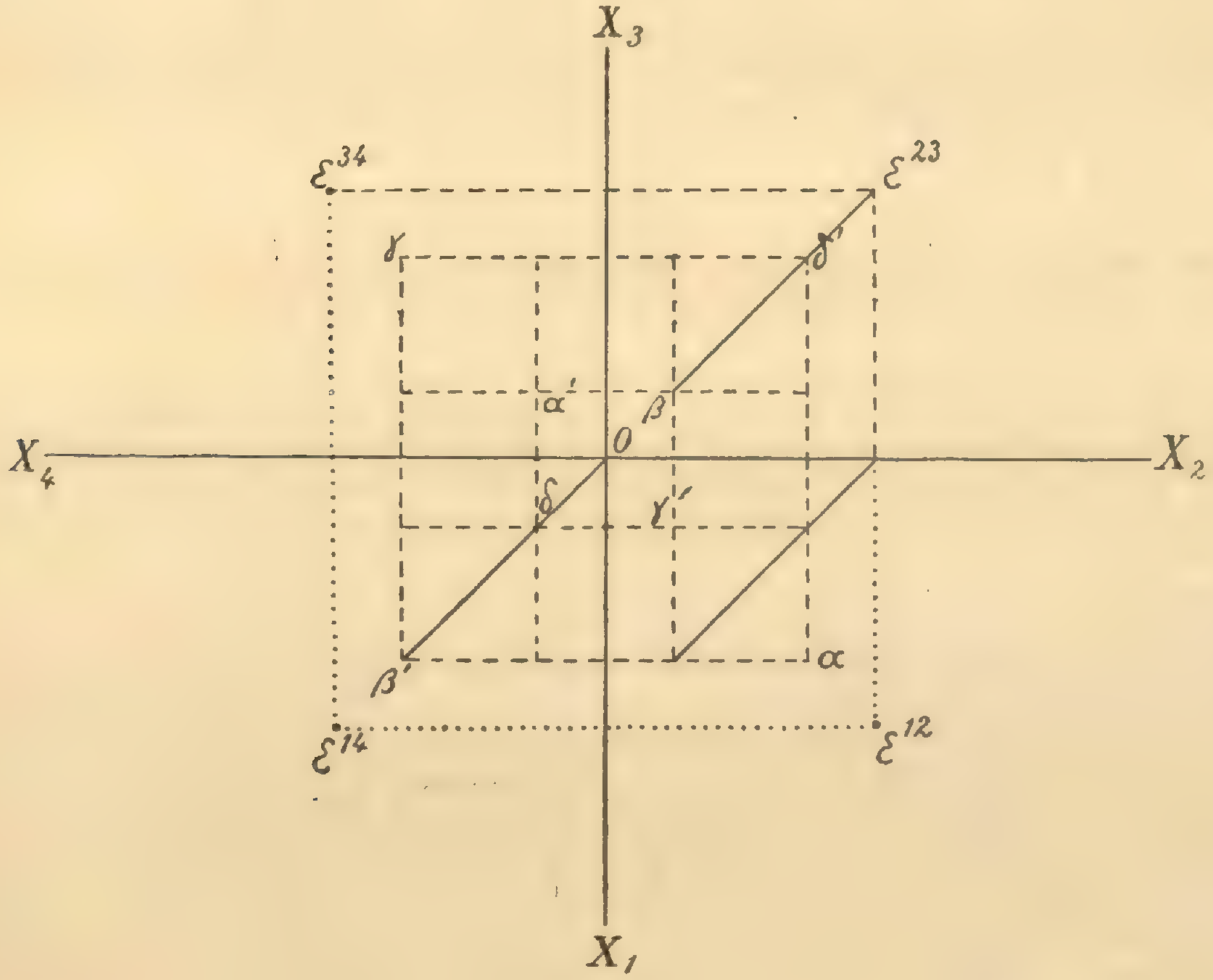

Fig. 2.

In these figures we have applied the methods of descriptive geometry (see Prof. SCHOUTE's *Lehrbuch der mehrdimensionalen Geometrie*) to obtain the completing fifths e, ε to the fours $abcd$, $\alpha\,\beta\,\gamma\,\delta$, starting from the property, that

in the double four of lines

$a\ b\ c\ d$
$a'\ b'\ c'\ d'$

the spaces (aa'), (bb'), (cc'), (dd') intersect in one line e, this being the completing fifth to either of the fours $a\,b\,c\,d$, $a'\,b'\,c'\,d'$.

in the double four of planes

$\alpha\ \beta\ \gamma\ \delta$
$\alpha'\ \beta'\ \gamma'\ \delta'$

the points α/α', β/β', γ/γ', δ/δ' lie in one plane ε, this being the completing fifth to either of the fours $\alpha\,\beta\,\gamma\,\delta$, $\alpha'\beta'\gamma'\delta'$.

We do not enter into the further constructions, though they are simple enough, it being rather our intention to generate these remarkable figures by constructions not going beyond our space, and basing ourselves on the more formal side of our correspondence.

We think it meet, to deal first with the following very simple problem:

There being given three couples of points a, b, c, to determine the incident couple l.

(Two couples of points are said to be incident if the pencils of x-spheres they determine have an element in common, in which case they lie on a circle).

There being given three circles α, β, γ, to determine the incident circle λ.

(Two circles, are said to be incident if they intersect in *two* real or imaginary points).

One easily recognizes in these propositions the transformed ones of the following:

To determine the transversal line l, to three given straight lines a, b, c.

To determine the transversal plane to three given planes α, β, γ.

The very simple solution of these may be represented symbolically thus:

$$(b\,c) = A \qquad \beta/\gamma = A$$
$$(c\,a) = B \qquad \gamma/\alpha = B$$
$$(a\,b) = \Gamma \qquad \alpha/\beta = C$$
$$A/B/\Gamma = l \qquad A\,B\,C = \lambda$$

Reading these symbols as follows:

ξ-sphere A through b and c
„ B „ c „ a
„ Γ „ a „ b
couple of points l, intersection of the three spheres A, B, Γ,

x-sphere A orthogonal to β and γ
„ B „ „ γ „ α
„ C „ „ α „ β
circle λ „ „ the three x-spheres A, B, C,

we have obtained a solution which we think to be the most natural one, even apart from our correspondence.

We now pass to the construction of the SEGRE configuration in our space Π.

We transform directly the theorems relative to this construction, they having been originally thought as theorems in S_4,

There being given four elements:

couples of points a, b, c, d | circles $\alpha, \beta, \gamma, \delta$

we can determine the incident element to every three of them, so that there be formed a double four

$$\begin{matrix} a & b & c & d \\ a' & b' & c' & d' \end{matrix} \quad \Big| \quad \begin{matrix} \alpha & \beta & \gamma & \delta \\ \alpha' & \beta' & \gamma' & \delta' \end{matrix}$$

in which any element is incident to the three not belonging to the same row or column.

We further determine

the four ξ-spheres:

$$(aa') \ , \ (bb') \ , \ (cc') \ , \ (dd') \ .$$

These intersect in one couple of points e.

the four x-spheres:

$$\alpha/\alpha' \ \beta/\beta' \ \gamma/\gamma' \ \delta/\delta'.$$

These admit one orthogonal circle ε.

If we apply this construction to any four of the set

$$a \ b \ c \ d \ e, \qquad \alpha \ \beta \ \gamma \ \delta \ \varepsilon,$$

it always leads back to the missing fifth.

Such five elements constitute

an *associated set of couples of points* | a STEFANOS *pentacycle*

The complete figure appearing in this way is the SEGRE *configuration.*

We finally propose another construction of the pentacycle and its dual figure, based upon a theorem of SEGRE which we reproduce here in its transformed form:

Six couples of points being given arbitrarily, there are 5 circles incident to each of these couples. They form a pentacycle.

Six circles being given arbitrarily, there are 5 couples of points incident to each of these circles. They form an associated set.

NOW, SCHUBERT's principle furnishes the following easy demonstration of the first part of this theorem, and, at the same time, another construction of these associated sets.

Considering the particular case, that the given couples of points aa' bb' cc' are incident two and two, so that

$$(aa') = \alpha \qquad a/a' = A$$
$$(bb') = \beta \qquad b/b' = B$$
$$(cc') = \gamma \qquad c/c' = B$$

the incident circles are:

$$\pi_1 = (A \ B \ C)$$
$$\pi_2 = (\beta/\gamma \ \gamma/\alpha \ \alpha/\beta)$$
$$\pi_3 = (A \ \beta)/(A\gamma)$$
$$\pi_4 = (B\gamma)/(B\alpha)$$
$$\pi_5 = (C\alpha/(C\beta)$$

These form a pentacycle.

Considering the particular case that the given circles $\alpha\alpha'$ $\beta\beta'$ $\gamma\gamma'$ are incident two and two, so that

$$\alpha/\alpha' = a \qquad (\alpha\alpha') = A$$
$$\beta/\beta' = b \qquad (\beta\beta') = B$$
$$\gamma/\gamma' = c \qquad (\gamma\gamma') = \Gamma$$

the incident couples of points are

$$p_1 = A/B/\Gamma$$
$$p_2 = (bc)/(ca)/(ab)$$
$$p_3 = (A/b \ A/c)$$
$$p_4 = (B/c \ B/a)$$
$$p_5 = (\Gamma/a \ \Gamma/b).$$

These form an associated set.

We leave the interpretation of these symbols to the reader.

Physics. — "*On the solid state.*" II. By J. J. VAN LAAR. (Communicated by Prof. H. A. LORENTZ.)

(Communicated in the meeting of April 23, 1909).

b. In the foregoing Paper [1]) the expression (see p. 770):

$$\frac{\beta^2}{1-\beta^2} = \frac{cT^{\gamma+1}\, e^{-\frac{q_0}{RT}}\, e^{-\frac{p+a/v^2}{RT}\Delta b}}{p+a/v^2} \quad . \quad . \quad . \quad . \quad (2)$$

was found for the dissociation of the double molecules of an arbitrary substance. With

$$\left.\begin{aligned} \frac{p+a/v^2}{RT}(-\Delta b) &= \frac{1+\beta}{v-b}(-\Delta b) = \varphi \\ \frac{RT}{q_0} &= \theta \quad ; \quad \frac{c\, q_0^{\gamma}}{R^{\gamma+1}}(-\Delta b) = \lambda \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad . \quad (\alpha)$$

(see p. 773) it was reduced to

$$\frac{\beta^2}{1-\beta^2} = \lambda\theta^{3/2}\, e^{-1/\theta}\, \frac{e^{\varphi}}{\varphi} \quad , \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

when $\gamma = 3/2$ was assumed. For as $\gamma = \frac{1}{R}(-k_1 + 2k_2)$, k_2 will be e. g. for a tri-atomic molecule about 6 (gr. cal.), and k_1 will be about 9 for the six-atomic double molecule. For γ we find then the value $1/2(-9+12) = 3/2$. But in other cases the exponent γ will, of course, possess other values. In order to be able to carry out some numerical calculations, we are at any rate justified in assuming an *imaginary* substance, the determining quantities of which are given by arbitrary, but possible values.

Thus we imagined a substance, for which (see p. 773)

$$\left.\begin{aligned} c = 2 \text{ (gr. cal., c.M.}^{-3}) \; ; \; q_0 = 3200 \text{ (gr. cal)} \; ; \; \gamma = 3/2 \\ a = 2700 \; ; \; b_1 = 1 \; ; \; 2b_2 = 1/2 \; ; \; -\Delta b = -(-b_1 + 2b_2) = 1/2 \end{aligned}\right\},$$

which caused equation (3) to pass into ($R = 2$ gr. cal.)

$$\frac{\beta^2}{1-\beta^2} = 32000\, \theta^{3/2}\, e^{-1/\theta}\, \frac{e^{\varphi}}{\varphi} \quad . \quad . \quad . \quad . \quad . \quad . \quad (3^a)$$

On the supposition a and b constant, the critical data of this substance will be determined by

$$2RT_c = \frac{8}{27}\frac{a}{b} \quad ; \quad p_c = \frac{1}{27}\frac{a}{b^2},$$

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[1]) These Proceedings, March 27th 1909, p. 765.

when we namely assume that for the critical point the dissociation of the double molecules has proceeded so far, that we may practically assume that all the molecules have become simple[1]). As a and b, as all the above quantities, refer to *double* molecular quantities, we must write $2RT_c$, because for $\beta=1$ the factor $1+\beta$ (in $(1+\beta)RT$) passes into 2.

Hence we find with $R=2$ (gr. cal.) and $b=2b_2={}^1/_2$:

$$4T_c=\frac{8}{27}\times\frac{2700}{{}^1/_2};\qquad p_c=\frac{1}{27}\times\frac{2700}{{}^1/_4},$$

or

$$T_c=400\ (\textit{absolute});\quad p_c=400.$$

But as we saw on p. 734 of our preceding paper, pv and ${}^a/_v$ are now expressed in caloric measure, as R is expressed in gr. cal. So to express these quantities in ergs, i. e. p and ${}^a/_{v^2}$ in dynes per cm^2, the found values of p and ${}^a/_{v^2}$ must still be multiplied by $41{,}74\times10^6$; or by 41,20, when p and ${}^a/_{v^2}$ are expressed in atmospheres.

As now a critical pressure of $400\times41{,}2=16480$ atms. is somewhat too high for an ordinary substance, we may obtain more suitable values of pressure without any change in our foregoing or following calculations by assuming all values of pressure to be e. g. 100-times smaller, and all values of volume 100-times larger, retaining all values of energy. So instead of $b_1=1\text{cm}^3$, $2b_2={}^1/_2\ \text{cm}^3$, $-\Delta b={}^1/_2\ \text{cm}^3$, we have only to think $b_1=100\ \text{cm}^3$, etc. In this case 270000 must be taken for a instead of 2700, for then the value of pressure ${}^a/_{v^2}$ becomes 100-times smaller than before, whereas the value of the energy ${}^a/_v$ remains the same.

Now for the low temperature $T=9$ (absolute), in consequence of which θ becomes $\frac{9}{1600}$ (see p. 774), (3^a) passes into

$$\frac{\beta^2}{1-\beta^2}=\frac{27}{2}\,e^{-\frac{1600}{9}}\frac{e^\varphi}{\varphi},$$

or

$$log^{10}\frac{\beta^2}{1-\beta^2}=-76{,}077+0{,}4343\,\varphi-log^{10}\varphi.\quad\ldots\quad(a)$$

Then the value of $p=\frac{(1+\beta)RT}{v-b}-\frac{a}{v^2}=\frac{2T}{-\Delta b}\varphi-\frac{a}{v^2}$ becomes:

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[1]) Otherwise the critical data will be determined by the equations (15), (16) and (17), derived by me in the Arch. Teyler (2) T. 11, Troisième partie, p. 235—331 (1909).

$$p = 36\varphi - \frac{2700}{v^2} \quad , \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (b)$$

whereas for $v = b + (v-b) = (b_1 + \beta\Delta b) + \frac{(1+\beta)(-\Delta b)}{\varphi}$ is found ($b_1 = 1, -\Delta b = {}^1/_2$):

$$v = 1 - {}^1/_2\left(\beta - \frac{1+\beta}{\varphi}\right), \quad . \quad . \quad . \quad . \quad . \quad . \quad (c)$$

which for values of β in the neighbourhood of 1 passes into $v = {}^1/_2 + \frac{1}{\varphi}$, and for values of β in the neighbourhood of 0 into $v = 1 + \frac{1}{2\varphi}$.

So we calculated the following values for β, v, and p for $T = 9$. To gain a better survey of the course of the values mentioned, some more values of φ have been inserted in the table.

We notice that for all the values of T, when viz. Δb is *negative*, i.e. φ *positive*, β has a *minimum value* at $\varphi = 1$. The amount of this minimum value will, of course, depend on the temperature. Thus $\beta_{min} = 1{,}5 \,.\, 10^{-38}$ or practically $= 0$ at $T = 9$. (For $v - b = 0$, $p = \infty$, β is always $= 1$, when Δb is negative, while for $v = \infty$, $p = 0$, β is also $= 1$).

When $\varphi = 1$, v is evidently $= 1{}^1/_2$ according to (c). In general v will be determined by $v = b_1 - \Delta b$ for $\varphi = 1$. (Comp. also p. 771 of I).

It is self-evident that for the accurate calculation of β, v and p more figures have been reckoned with than are indicated here. The values of β and v have mostly been shortened to 2 figures, whereas those of p have been indicated in 2 or 3 figures. So between $\varphi = 187$ and $\varphi = 0{,}25$ the values of p have been reduced to round numbers to tens of units.

With regard to the different maxima and minima e.g. for $T = 9$, we observe once more, that in the equation:

$$p = \frac{(1+\beta)RT}{v-b} - \frac{a}{v^2} = \frac{RT}{-\Delta b}\varphi - \frac{a}{v^2},$$

in which a decrease of φ is always attended with an increase of v [1]), the pressure p first decreases rapidly from ∞ to -3840, when

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[1]) For from $v = b_1 - (-\Delta b)\left(\beta - \frac{1+\beta}{\varphi}\right)$ follows:

$T = 9.$

$\varphi = \frac{1+\beta}{v-b}(-\Delta b)$	$log^{10}\ \frac{\beta^2}{1-\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
300	51,734	$5{,}42.10^{51}$	$1-\frac{1}{1{,}08\ 10^{52}}$	0,503	+36
187	2,864	731	$1-\frac{1}{1460}$	0,506	—3830
186	2,432	270	$1-\frac{1}{540}$	0,506	—3840
185	1,999	99,8	0,995	0,508	—3810
183	1,137	13,7	0,97	0,52	—3230
182	0,705	5,07	0,91	0,55	—2440
181	0,273	1,87	0,81	0,60	—960
180	— 0,160	0,692	0,64	0,68	+720
179	— 0,591	0,256	0,45	0,78	+1980
178	— 1,023	0 0948	0,29	0,86	+2730
177	— 1,455	0,0351	0,18	0,91	+3130
175	— 2,318	0,00481	0,069	0,97	+3420
173	— 3,182	0,000658	0,026	0,99	+3470
170	— 4,478	0,0000333	0,0058	1,000	+3420
100	—34,648	$2{,}25.10^{-35}$	$4{,}74.10^{-18}$	1 005	+930
50	—56,061	$8{,}69.10^{-57}$	$9{,}32\ 10^{-29}$	1,01	—850
10	—72,734	$1{,}85.10^{-73}$	$4{,}30.10^{-37}$	1,05	—2090
8	—73,506	$3{,}12.10^{-74}$	$1{,}77.10^{-37}$	1,06	—2100
5	—74,605	$2{,}48.10^{-75}$	$4{,}98.10^{-38}$	1,10	—2050
1	—75,643	$2{,}28.10^{-76}$	$1{,}51.10^{-38}$ (min.)	1,5	—1160
0,5	—75,559	$2{,}76.10^{-76}$	$1{,}66.10^{-38}$	2,0	—660
0,25	—75,366	$4\ 31.10^{-76}$	$2{,}08.10^{-38}$	3,0	—290
0,1	—75,034	$9{,}25.10^{-76}$	$3{,}04.10^{-38}$	6,0	—71
0,01	—74 073	$8{,}45.10^{-75}$	$9{,}19.10^{-38}$	51	— 0,68
0,0025	—73,474	$3{,}36.10^{-74}$	$1{,}83\ 10^{-37}$	200	+ 0,022
0,0017	—73,306	$4{,}94\ 10^{-74}$	$2{,}22.10^{-37}$	300	+ 0,030
0,0010	—73,077	$8{,}38\ 10^{-74}$	$2{,}89.10^{-37}$	500	+ 0,025
10^{-6}	—70,077	$8{,}38.10^{-71}$	$9{,}15.10^{-36}$	$0{,}50.10^{6}$	36.10^{-6}
10^{-74}	— 2,077	0,00838	0,091	$0{,}55.10^{74}$	36.10^{-74}
10^{-75}	— 1,077	0,0838	0,28	$0{,}64\ 10^{75}$	36.10^{-75}
10^{-76}	— 0,077	0,838	0,68	$0{,}84.10^{76}$	36.10^{-76}
10^{-77}	0,923	8,38	0,92	$0{,}96.10^{77}$	36.10^{-77}
0	∞	∞	1	∞	0

φ decreases from ∞ to 186. As, namely, here v remains nearly constantly $= {}^1/_2$ (β remains practically $= 1$), ${}^a/_{v^2}$ will hardly change at all, so that the whole decrease of p is practically caused by the decrease of φ, i.e. by the increase of the small value of $v-b$ [in consequence of the (slight) increase of v]. But in the neighbourhood of the minimum E, β will begin to change rapidly, and decrease from 1 to about 0 in the neighbourhood of the maximum D, on account of which the value of v will increase as rapidly from ${}^1/_2$ to 1, so that ${}^a/_{v^2}$ decreases from 10800 to 2700. Now in consequence of this rapid decrease of ${}^a/_{v^2}$, while φ changes little (at $T = 9$ from 186 to 173)[1]), p will increase rapidly between E and D (see the Plate in the preceding paper). Between D and C we have again the same thing as between $p = \infty$ and the point E: β and v remain namely almost unchanged (resp. $= 0$ and 1), so that now for decreasing φ the value of ${}^a/_{v^2}$ changes again little, and p will only decrease through the decrease of φ (from 173 to 8); i.e. through the increase of the slight value of $v-b$ in consequence of the (slight) increase of v. As we know, the increase of $v-b$ will be less between C and B than the decrease of ${}^a/_{v^2}$ in consequence of the great increase of v, so that p rises; while past B the further decrease of $v-b$ or of v predominates, as then ${}^a/_{v^2}$ may be neglected by the side of $\frac{RT}{v}$.

$$-\frac{dv}{d\varphi} = (-\Delta b)\left(\frac{1+\beta}{\varphi^2} + \left(1-\frac{1}{\varphi}\right)\frac{d\beta}{d\varphi}\right).$$

Further by logarithmical differentiation we find from $\frac{\beta^2}{1-\beta^2} = A\frac{e^\varphi}{\varphi}$:

$$\frac{d\beta}{d\varphi} = \frac{\beta(1-\beta^2)}{2}\left(1-\frac{1}{\varphi}\right),$$

so that we find for $-\frac{dv}{d\varphi}$:

$$-\frac{dv}{d\varphi} = (-\Delta b)\left(\frac{1+\beta}{\varphi^2} + \frac{\beta(1-\beta^2}{2}\left(1-\frac{1}{\varphi}\right)^2\right),$$

which is always positive.

[1]) On p. 780 (Cf. also p. 766) of the preceding paper the temporary decrease of $v-b$ has been erroneously given as the cause. The small value of $v-b$ does, indeed, decrease between E and D, where both v and b rapidly increase — but $1+\beta$ decreases in about the same degree, so that the fraction $\frac{1+\beta}{v-b} = \frac{\varphi}{-\Delta b}$ changes but little. It is merely the rapid decrease of ${}^a/_{v^2}$, to which the increase of the pressure between E and D is owing.

We add that in I (p. 779) the value $\beta = 1 - \frac{1}{600}$ was found in the minimum E, which corresponds with a value of φ somewhat greater than 186. In the point D we found $\beta = 0{,}0267$, to which belongs a value of φ only little higher than 173. As in the points C and B the values of v were found resp. 1,063 and 298, the corresponding values of φ are resp. somewhat smaller than 8 and somewhat larger than 0,0017.

8. In this we meet with an important question. Towards what limiting values will these maxima and minima converge, when T approaches to 0?

Though, namely. the possibility exists, that for exceedingly low temperatures factors come into play, which have not been reckoned with in our equations, yet it does not seem to me to be devoid of importance, to ascertain what follows from these equations for the limiting case $T = 0$.

In the equation (3), viz.:

$$\frac{\beta^2}{1-\beta^2} = \lambda \frac{\theta^{3/2}}{\varphi} e^{\varphi - 1/\theta},$$

β will soon approach 1 for values of $\varphi > \frac{1}{\theta}$, in consequence of the great increase of the exponential factor; while for values of $\varphi < \frac{1}{\theta}$, for which the exponent becomes negative, β will rapidly approach to 0.

The great change of β from 1 to 0 between E and D, which determines the minimum at E and the maximum at D, takes therefore place for values of φ, which lie in the neighbourhood of $\frac{1}{\theta}$.

For $T = 9°$ we have $\frac{1}{\theta} = \frac{1600}{9} = 178$, and now we really found $\varphi_E = 186$ and $\varphi_D = 173$. As now according to (α)

$$\varphi - \frac{1}{\theta} = \frac{(p + a/v^2)(-\Delta b) - q_0}{RT},$$

we shall evidently have $\varphi_E = \varphi_D = \infty$ for $T = 0$, and this value will be determined by $\varphi = \frac{1}{\theta}$, so that we have:

$$(T = 0) \quad p_{E,D} = \frac{q_0}{-\Delta b} - \frac{a}{v^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

For in consequence of $T = 0$ the exponent $\varphi - \frac{1}{\theta}$ will now have the value $+\infty$ ($\beta = 1$) for $(p + {}^{a}/_{v^2})(-\Delta b) > q_0$, and for id. $< q_0$ the value $-\infty$ ($\beta = 0$). Only when $(p + {}^{a}/_{v^2})(-\Delta b) = q_0$, β can have a value between 1 and 0 (the portion ED), as then the exponent $\varphi - \frac{1}{\theta}$ assumes the indefinite value $\frac{0}{0}$.

So in our example, where $q_0 = 3200$, $-\Delta b = {}^{1}/_{2}$, $a = 2700$, we shall find in E, where $v = 2b_2 = {}^{1}/_{2}$:

$$(T = 0) \quad p_E = 6400 - 10800 = -4400,$$

whereas we find in D_1, where $v = b_1 = 1$:

$$(T = 0) \quad p_D = 6400 - 2700 = 3700.$$

So these are the limiting values to which the pressure in E and D approaches when T gets near 0. For $T = 9$ these values are resp. -3840 and $+3470$.

The fact now that one value is *negative*, the other *positive*, so that a *positive* pressure of coexistence is possible, is determined by this, that somewhere between D and E p can become $= 0$, in other words that q_0 lies between $\frac{a}{(2b_2)^2}(-\Delta b)$ and $\frac{a}{b_1^2}(-\Delta b)$. In our example this is the case, as $3200 < 5400$ and > 1350.

If $q_0 < \frac{a}{b_1^2}(-\Delta b)$, so this would be < 1350 in our case, also p_D is negative, and then a coexistence liquid-solid would only be possible for negative pressures — in other words, *no solid state* would appear in this case.

If, however, $q_0 > \frac{a}{2b_2^2}(-\Delta b)$, i. e. > 5400 in our case, P_E and P_D are both *positive*, and then a solid state is possible for higher pressures (also in the triple point).

We have tacitly assumed in the above that Δb is *negative*, so $-\Delta b$ positive. If, however, Δb is *positive*, which case we shall treat separately later on, the factor $\frac{e^{\varphi}}{\varphi}$ in the second term of (3) passes into $\frac{e^{-\varphi}}{\varphi}$ according to (2) and (α) (when we namely then put $\frac{1+\beta}{v-b}\Delta b = \varphi$), and then β will increase from 0 to 1 without minimum, if v increases from b_1 to ∞.

It is, however, possible, that in this case β increases so rapidly

already between b_1 and $2b_2$ that here too a maximum at D occurs at a suitable place and a minimum at E. As appears from (3) or (3^a), this will, of course, only be the case when the temperature rises above a certain value (in the neighbourhood of the triple-point temperature), just as for Δb negative the temperature must descend below a certain value to bring this about. (See also fig. 4 of the plate in the foregoing paper, in which figure the line SM will now run to the right).

But as has been said, we shall discuss these relations — specially in reference to the situation of the triple point and the course of the line SM — more fully in a subsequent paper both in the case Δb positive and in the case Δb negative.

Let us now examine the limiting values of the minimum at C and the maximum at B, when T converges to 0. As past D the change of β (from 1 to 0) has come to an end, if $T=0$, $\frac{dp}{dv}$ can be found from $p=\frac{RT}{v-b_1}-\frac{a}{v^2}$, yielding $\frac{dp}{dv}=\frac{2a}{v^3}-\frac{RT}{(v-b_1)^2}$. So this is 0, when

$$\frac{(v-b_1)^2}{v^3}=\frac{RT}{2a},$$

so that then either $v=b_1$ or $v=\infty$ for $T=0$.

If we substitute $(v-b_1)^2=\frac{RTv^3}{2a}$ in the equation for p, we get:

$$p=\sqrt{\frac{2a\,.\,RT}{v^3}}-\frac{a}{v^2},$$

which for $T=0$, $v=b_1$ leads to:

$$(T=0)\quad p_C=-\frac{a}{b_1^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

Hence to -2700 in our example. (For $T=9$ we found -2100). Then the value of $\varphi=\frac{1+\beta}{v-b}(-\Delta b)$ is $=\infty$. (For $T=9$ we had $\varphi_C=8$, so that φ_C rapidly increases, when T comes near 0).

If $v=\infty$, p is evidently $=0$. Then φ is also $=0$. [1])

[1]) If at low temperature $\beta=0$ in C and B, then approximately

$$v_C-b_1=\sqrt{\frac{Tb_1^3}{a}}\quad;\quad v_D=\frac{a}{T}$$

follows from $\frac{(v-b_1)^2}{v^3}=\frac{RT}{2a}$.

So with our values for $T=9$ $v_C=1+\sqrt{\frac{1}{300}}=1{,}06$ and $v_D=300$.

For φ we find:

Graphically the isotherm $T = 0$ is indicated by the straight line FE, i. e. the vertical axis $v = 2b_2\,(\beta = 1)$; the curve ED along which β changes from 1 to 0; the straight line DC, i. e. the vertical axis $v = b_1\,(\beta = 0)$; and finally the horizontal straight line CB, which does not meet the horizontal axis $p = 0$ before $v = \infty$.

The direction of the curve ED is found from the general equation (7) of Part I (p. 778), by putting $\varphi = \infty$ (see above). (so $v - b$ is all through the change from $\beta = 1$ to $\beta = 0$ constantly $= 0$, hence $v = b$). Then we get:

$$\frac{dp}{dv} = \frac{2a}{v^3} - \frac{(1+\beta)\,RT}{(v-b)^2}\,\frac{1}{{}^1/_2\beta\,(1-\beta)\,\varphi^2}\,,$$

or as $(v-b)^2\varphi^2 = (1+\beta)^2\,(-\Delta\,b)^2$:

$$\frac{dp}{dv} = \frac{2a}{v^3} - \frac{2RT}{\beta(1-\beta^2)\,(-\Delta b)^2}\,.$$

So along ED we have for $T = 0$:

$$\frac{dp}{dv} = \frac{2a}{v^3}\,,$$

hence:

$$\left(\frac{dp}{dv}\right)_E = \frac{2a}{(2b_2)^3}\quad;\quad \left(\frac{dp}{dv}\right)_D = \frac{2a}{{b_1}^3}\,.$$

As now (see above):

$$p_D - p_E = \frac{a}{(2b_2)^2} - \frac{a}{{b_1}^2},$$

the curve ED will rise more rapidly at E than the connecting line ED, and less rapidly at D.

Since according to equation (8) the limiting pressures p_E and p_D will approach to *finite* values for $T = 0$, the coexistence pressure solid-liquid, which at all events must lie between p_E and p_D, must also possess a *finite* value for $T = 0$, so that in Fig. 4 of the plate of Part I the line SM cannot under any circumstances ascend to

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$$\varphi_C = (-\Delta b)\sqrt{\frac{a}{Tv^3}}\quad;\quad \varphi_D = (-\Delta b)\,\frac{T}{a}.$$

So at $T = 9$ for our substance $\varphi_C = {}^1/_2\sqrt{252} = 7{,}9\quad;\quad \varphi_D = \frac{1}{600} = 0{,}0017.$

And for p we may write:

$$p_C = \sqrt{\frac{4aT}{v^3}} - \frac{a}{v^2}\quad;\quad p_D = \frac{2T^2}{a} - \frac{T^2}{a} = \frac{T^2}{a},$$

which yields $p_C = 290 - 2400 = 2110\quad;\quad p_D = 0{,}03$ for $T = 9$.

All these values harmonize perfectly with those found in the table.

infinity, as was formerly sometimes supposed, but it will indicate a very decided (positive or negative) value of the pressure for $T=0$. Hence with *high pressure* we must always reach the liquid state through the solid state, when Δb is *negative*, however low the temperature may be. And in the same way the *solid* state through the liquid state, when for Δb *positive* (*SM* then runs to the *right*) the temperature gets above the triple-point temperature.

But we shall soon discuss this more fully. We may now only add the following remarks to what precedes.

As $p = \frac{q_0}{-\Delta b} - \frac{a}{v^2}$ everywhere between E and D for $T=0$ (see above), we find the pressure of coexistence solid-liquid p_0 evidently from

$$p_0 = \frac{1}{v_1 - v_2} \int_{v_2}^{v_1} \left(\frac{q_0}{-\Delta b} - \frac{a}{v^2} \right) dv,$$

yielding:

$$(T=0) \qquad p_0 = \frac{q_0}{-\Delta b} - \frac{a}{v_1 v_2} \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

In this v_1 is the volume of the solid phase ($= b_1$) and v_2 that of the liquid phase ($= 2b_2$), while $v_1 - v_2 = -\Delta b$. With our values this becomes therefore $p_0 = 6400 - 5400 = 1000$, so a positive pressure.

The course of β along the curve DE may be found from:

$$v = b = b_1 - \beta(-\Delta b),$$

from which:

$$(T=0) \qquad \beta = \frac{b_1 - v}{-\Delta b}, \quad . \quad . \quad . \quad . \quad . \quad . \quad (11)$$

running from 1 at $v = 2b_2$ to 0 at $v = b_1$.

The value of v_0 on the ideal isotherm ED, corresponding with the pressure of coexistence p_0, is found from $p = \frac{q_0}{-\Delta b} - \frac{a}{v^2}$, yielding:

$$\frac{q_0}{-\Delta b} - \frac{a}{v_1 v_2} = \frac{q_0}{-\Delta b} - \frac{a}{v_0^2},$$

hence

$$v_0 = \sqrt{v_1 v_2}.$$

As to the direction of the p,T-line SM for $T=0$, it can be determined from the well known equation:

$$\frac{dp}{dT} = \frac{\Delta E}{T \Delta V}.$$

In this $\Delta E = E_{liquid} - E_{solid}$. As now in general:

$$E = [(1-\beta)(e_1)_0 + 2\beta(e_2)_0] + [(1-\beta)k_1 + 2\beta k_2]\,T - \frac{a}{v} + pv,$$

in other words (as $-(e_1)_0 + 2(e_2)_0 = q_0$ and $k_1 + 2k_2 = \gamma R$):

$$E = [(e_1)_0 + \beta q_0] + (k_1 + \beta\gamma R)\,T - \frac{a}{v} + pv,$$

we have (the index 2 refers to the liquid phase):

$$\Delta E = (\beta_2 - \beta_1)(q_0 + \gamma RT) - \left(\frac{a}{v_2} - \frac{a}{v_1}\right) + p(v_2 - v_1).$$

In this $v_2 - v_1 = \Delta V$, hence we get generally:

$$\Delta E = (\beta_2 - \beta_1)(q_0 + \gamma RT) + \left(p + \frac{a}{v_1 v_2}\right)\Delta V. \quad . \quad . \quad . \quad (12)$$

Now for $T=0$ we have $p = p_0 = \frac{q_0}{-\Delta b} - \frac{a}{v_1 v_2}$, $\Delta V = 2b_2 - b_1 = \Delta b$,

so that only remains:

$$(T = 0) \qquad \Delta E = \gamma RT. \quad . \quad . \quad . \quad . \quad . \quad . \quad (12^a).$$

So finally we get for $T = 0$:

$$(T = 0) \qquad \left(\frac{dp}{dT}\right)_0 = \frac{\gamma RT}{-T(-\Delta b)} = -\frac{\gamma R}{-\Delta b} \quad . \quad . \quad . \quad (13)$$

So also this limiting value is finite, so that for $T = 0$ the line SM meets the vertical axis $T = 0$ at a *finite* pressure and a *finite* angle. With our values $\left(\frac{dp}{dT}\right) = -\frac{3}{1/2} = -6$.

It follows from (10) that p_0 is *positive* only when $q_0 > \frac{a}{v_1 v_2}(-\Delta b)$.

If q_0 is precisely equal to this value, then $p_0 = 0$; and if q_0 is less than this value, p_0 would become negative, which is impossible. In our example the limiting value meant is 2700.

It is self-evident that in this Δb must always be negative. For otherwise the line SM does not run to the left, but to the right, and coexistence solid-liquid is not found below the triple-point temperature, but is found at higher temperatures.

9. In connection with the foregoing considerations it has sometimes been asked why the new phase, the existence of which — in consequence of the renewed bent of the isotherms in the neighbourhood of $v = b$ on the assumption of *association, when* Δb *differs from zero — is undeniable*, would have to be a *solid* phase. Can it not be a *second liquid* phase, so that a "longitudinal plait" occurs with corresponding imperfect miscibility of two liquids?

To this we answer that a real longitudinal plait is only possible for a mixture of two *different* substances which are *not* in internal equilibrium with each other, as e. g. for phenol and water. For then there is for a *series* of temperatures "three-phase-equilibrium" possible (liquid I, liquid II, vapour). For if we denote these three phases by A, B, and C, we have $(\mu_1)_a = (\mu_1)_b = (\mu_1)_c$; $(\mu_2)_a = (\mu_2)_b = (\mu_2)_c$ for the three-phase-equilibrium, i.e. four independent relations between the 5 quantities p, T, x_a, x_b, x_c, so that *for any arbitrarily chosen value* of T the pressure (vapour pressure) p, as well as the three concentrations x_a, x_b, and x_c are entirely determined. We know that then the temperature at which three-phase-equilibrium is possible, ranges from the lowest temperatures to the so-called critical temperature of complete mixture, where two phases become identical.

In our case, however, it is different. In consequence of the internal equilibrium between the two kinds of molecules (simple and complex ones) we have not to deal with *two* substances in the sense of the theory of phases, but only with one substance, of which the internal molecular state is entirely determined in consequence of the equilibrium of association. Here only three-phase-equilibrium is possible at *one* temperature (the so-called "triple point") — in perfect accordance with the ordinary scheme for a simple substance. (see fig. 4 of the plate of part 1 of this paper). For now for three-phase-equilibrium we have the relations $(\mu_1)_a = (\mu_1)_b = (\mu_1)_c$ between the molecular potentials of the 1st component (e.g. the complex molecules), but at the same time in *each* of the phases $\mu_1 = \mu_2$, the relation of equilibrium between the two kinds of molecules. Then it follows naturally that $(\mu_2)_a = (\mu_2)_b = (\mu_2)_c$ is also fulfilled for the 2nd component. So there are here 5 independent relations (viz. the two relations $(\mu_1)_a = (\mu_1)_b = (\mu_1)_c$ and the three relations $(\mu_1)_a = (\mu_2)_a$, $(\mu_1)_b = (\mu_2)_b$, $(\mu_1)_c = (\mu_2)_c$) between the 5 quantities p, T, β_a, β_b, and β_c, so that for three-phase-equilibrium nothing remains indefinite, and besides the three degrees of dissociation β_a, β_b, and β_c also the (vapour) pressure and the temperature are perfectly determined.

The phenomenon studied by us agrees, therefore, entirely with the appearance of a *triple point* for a *simple* substance at *one definite* temperature, and had nothing to do with the appearance of a longitudinal plait for a binary mixture, for which three-phase-equilibrium is possible for a *series* of temperatures. So there is no question of real so-called imperfect miscibility. For a binary mixture the concentration x may be changed *at pleasure*; for a simple substance with internal equilibrium we cannot change β arbitrarily. For an ordinary binary mixture two different phases appear on imperfect miscibility

of the mixture (in consequence of changes of the values of x or T), which phases *differ* in concentration etc. from the original homogeneous phase; for a simple phase with internal equilibrium a new phase simply exists *by the side* of the original phase, e. g. a solid phase by the side of a liquid one, and that only with change of the temperature (or of the pressure). And these two phases can only coexist at the transition point, not with a series of concentrations x of the original homogeneous phase, as in the case of the ordinary imperfect miscibility. As soon as the temperature (or the pressure) differs however slightly from that of the transition point, one of the two phases *vanishes* at once.

And this, we think, sufficiently refutes the "longitudinal plait argument" raised by some.

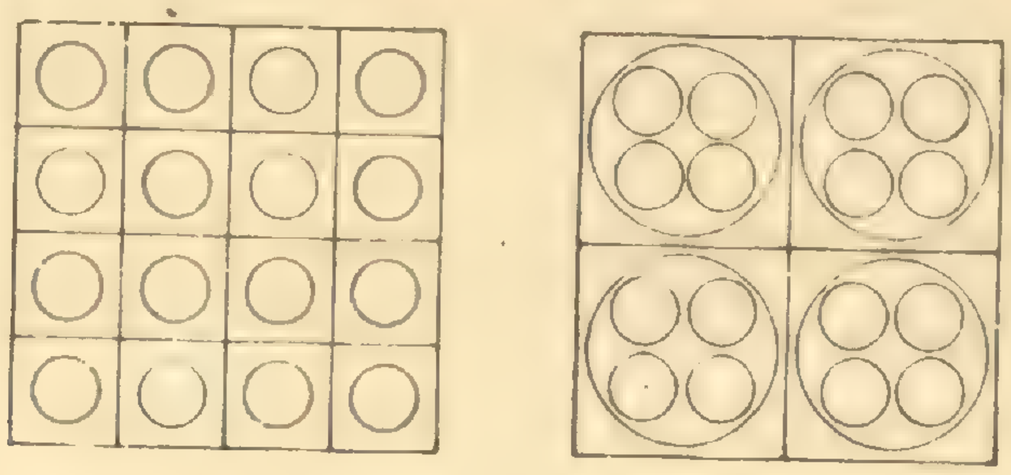
So a new (third) phase for a simple substance — and this phase can hardly be another than the third phase, which we know for nearly all simple substances: the *solid* one. Below the triple-point temperature it can either be in equilibrium with the vapour or with the liquid (for high pressures, if Δb is negative); only in the triple point it can be in equilibrium both with the vapour and the liquid.

But why does the new phase possess the properties of the "solid" state?

We first put the question: Why does the second phase, which may exist by the side of the gaseous one in the ordinary equation of state of VAN DER WAALS, possess the properties of the "liquid" state? Because its density is so much *greater*. And from this ensue all the properties which we observe in the liquid state. These properties cannot be directly derived from the equation of state itself: the fact of the greater internal friction e. g. follows only from the smaller mutual distance of the molecules; it is not to be derived from the equation of state, which remains very simple. Also the internal friction for gases can only be derived from kinetic considerations in a very intricate way, and it is really not to be deduced from the exceedingly simple relation $pv = RT$!

And now the answer to our first question. The third phase is *solid*, because in the formation of larger molecule-complexes the *mobility* decreases to such a degree that the molecules no longer can pass over and past each other as in the liquid state, but will necessarily be fixed in their positions. The fact of this slighter mobility is quite independent of the value of Δb, which is mostly very slight. We have namely exclusively to deal with the fact, that while the *total volume remains almost unchanged*, about the same intramolecular space $v-b$ is found both for the smaller simple molecules and for

the so much larger complex molecules so that the *ratio* between the diameter of a complex molecule and the linear dimension of the surrounding space is *many times smaller* than the same ratio for the simple molecules. A single glance at the adjoined schematical repre-



simple molecules (liquid)

quadruple molecules (solid)

sentation makes this at once clear. In the lefthand figure the *relative* linear extent of the interval is relatively large with respect to the *diameter* of the molecules; in the righthand figure it is comparatively small on the other hand. In other words: the complex molecules of the righthand figure can no longer move past each other: the molecule is too large to do so compared with the intervals; it will now be confined to a fixed centre of equilibrium, round which it can only move in closed paths; whereas in the lefthand figure the mobility is perfectly secured [1]).

The transition of the liquid to the solid state is, accordingly, brought about in consequence of this, that the *solid* state is characterized by extensive *formation of complexes* of the molecules; not only *two* simple molecules join to one double molecule, but possibly ten or a hundred to one multiple molecule, quite in accordance with the assumption of complex "crystal molecules".

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[1]) As for liquids and solid bodies under ordinary pressures p may be neglected by the side of a/v^2; the expression $v - b = \frac{(1+\beta)RT}{a/v^2}$ follows for $v - b$ from the equation of state, hence the expression $\frac{RT}{a} \times (1+\beta)\, v$ for $\theta = \frac{v-b}{v}$. Now in most cases v_2 (liquid) will differ but little from v_1 (solid), so the ratio θ will be about proportional to $1+\beta$; for multiple molecules with $1+(n-1)\,\beta$. So if in the liquid state $\beta_2 = 1$, in the solid state $\beta_1 = 0$, we shall have:

$$\theta_1 : \theta_2 = 1 : n,$$

so that in the "solid" state the ratio of the intra-molecular intervals to the whole volume, which ratio may to a certain extent be taken as a measure for the mobility, will be about n-times smaller than the corresponding ratio in the liquid state.

That viscosity strongly increases with association is a well-known fact. See e.g. BINGHAM, Viskosität und Fluidität, Z. f. ph. Ch. **66**, p. 245.

For a *slight* number of simple molecules could very well form a multiple molecule in some cases, without it being necessary that the phase becomes solid. For in all the so-called associating liquids a comparatively smaller or larger number of double, at most triple molecules are assumed.

In the theory worked out by us we assumed for the sake of simplicity only the formation of double molecules; we observed, however, already at the conclusion of our preceding paper that qualitatively the theory is not subjected to any change when multiple molecule complexes are started from. Still, I intend to revert to this later on.

With regard to the *number* of simple molecules which contribute to the formation of a multiple molecule, it is still worthy of note, that of course, this number may be different for one and the same substance according to circumstances. This explains too that several solid modifications often occur for one substance, which may pass into each other under certain circumstances and two or three of which — according to the phase rule — can exist by the side of each other, both with simultaneous coexistence of liquid and vapour or not.

That the properties of the solid state depend on the *direction*, is also a consequence of the relatively (with respect to the diameter of the molecule) slighter distances, and the being bound to fixed centres of equilibrium, in consequence of which also the grouping of the atoms in the molecule can come more to the fore. But on the whole it will after all be probably the grouping of the simple molecules inside the complex crystal molecules, so the structure of the last-named, which in perfect accordance with the prevailing crystallographic ideas, causes the properties to change with the direction.

Strictly speaking the equation of state will be different there for every direction, because the specific quantities $\sqrt[3]{v}$ and $\sqrt[3]{b}$ vary with the direction. But yet we may treat v and b, like a, as scalar quantities, when only here as elsewhere, we have in view *middle values*. Then there is no reason why we should not continue to speak of a definite specific volume v, or density $1/v$, or of a definite molecular volume b in these cases, just as we can continue to speak of a definite specific attraction a. For all the so-called optically-isotropic solid phases, moreover, the difference of direction disappears for the different properties.

So we can summarize the above considerations as follows:

For a simple substance, the molecules of which can combine to larger complexes, and where Δb differs from zero, so that e. g. for

negative Δb the complex-formation again greatly decreases in the neighbourhood of $v = b$ in consequence of increase of pressure (or diminution of volume), the following possibilities will exist:

a. a phase with large specific volume (*slight density*): a *gasphase.*

b. a phase with small specific volume (*great density*), but internally still sufficiently mobile in consequence of comparatively slight complex formation: a *liquid phase.*

c. a phase also with *great density*, but internally *immobile* in consequence of extensive complex formation: a *solid phase.*

For negative Δb the density of the solid phase is even less [1]) than that of the liquid phase; it is however not so much the density, but the formation of the large complexes, which makes the phase immobile — as we have set forth fully above.

And just as there are all kinds of transitions possible between gas and liquid phases, and often no external difference is noticeable between the two phases, e.g. in the neighbourhood of the critical temperature, somewhat below it, or for a gas phase under high pressure above the critical temperature — in the same way there will of course be transitions between liquid and solid phases: liquid phases with relatively slight mobility (so-called amorphous-glassy state) and solid phases with comparatively great mobility (e.g. the fluid crystals or anisotropic liquids).

The real solid crystalline state is always, just as the corresponding liquid-crystalline state, connected with extensive complex formation; whereas the ordinary liquid state, together with its viscous continuation to the internally immobile amorphous-glassy state will be characterized by comparatively slight complex formation. In the last named state only the comparatively low temperature in connection with the greater density and the comparatively large volume of the molecules causes the immobility, the fixedness of the phase.

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[1]) For then Δv is generally also negative. Yet we must not forget that Δb and Δv generally differ. On p. 8 of my paper in the Arch. Teyler I derived the relation:

$$\Delta v = \frac{\Delta b + \frac{v-b}{1+\beta}}{1 - \frac{2a}{v^3}\,\frac{v-b}{p+a/v^2}}.$$

As we see, only for very small $v-b$ (compared with Δb) Δv is approximately $= -\Delta b$. But if $v-b$ is comparatively large with respect to Δb, Δv may even be *positive*, even if Δb is negative. We need only think of the gaseous state, where Δb is negligible by the side of $v-b$, and where Δv is simply $= \frac{v-b}{1+\beta} = \frac{v}{1+\beta}$.

It has been assumed by me in the above that the ordinary shape of VAN DER WAALS' equation of state remains also valid when the substance assumes the "solid" state in consequence of other molecular groupings. I do not shut my eyes to the fact that there are still some difficulties left, particularly with respect to the special forces which possibly come into play, and which possibly may modify the figure given by me in some respects; but I yet believe that through my considerations at least some insight is gained into the origin of the *third* (solid) phase, enclosed in the equation of state in its ordinary shape, when the association of the molecules is taken into account and the variation of volume attending it.

Chemistry. — "*On a synthesis of aldehydes and indole* [3]." By Dr. R. A. WEERMAN. (Communicated by Dr. W. A. VAN DORP).

(Communicated in the meeting of April 23, 1909).

Phenylacetaldehyde.

In a previous communication [1]) a method was described for the preparation of substances of the structure:

$$C_6H_5\text{-}C = C - N - C.$$

It now appeared that by the action of acids similar compounds yield aldehydes with destruction of the bond between the nitrogen and the double bound carbon. In this way methyl styrylaminoformate generates phenylacetaldehyde.

$$C_6H_5C^H = C^H - N^H - C^O_{OCH_3} \rightarrow C_6H_5C^H = C^H_{OH} + NH_3 + CO_2 + CH_3OH$$

$$\downarrow$$

$$C_6H_5C^{H_2} - C^H_O.$$

The oxime melts at 99°—100°

0,2115 gr. yielded 19,2 cc. N_2 at 759 mM. and 13°

Found: 10,66% N

Calculated for C_8H_9NO: 10,37% N.

Orthonitrophenylacetaldehyde. [2])

This aldehyde is generated in an analogous manner from methyl

[1]) These Proc. 1907.

[2]) Compare MOHR Journ. f. Pr. Ch. [2] 75, 549.

[3]) In view of a recently published article by SEMMLER (Ber. 42, 584), in which he shows that phenylacetaldehyde can readily pass into the phenylvinylalcohol, there is every reason for assuming that orthonitrophenylacetaldehyde may also react tautomerically and in connection with the negative substituent, it is even not impossible that in the solid condition it possesses the vinyl alcohol structure.

orthonitrostyrylaminoformate (see last communication). The b.p. is 133°—135° at 5 mm. the melting point 22°—23°.

0,1450 gr. yielded 0,3081 gr. CO_2 and 0,0546 gr. H_2O

0,1591 gr. yielded 11,9 cc. N_2 at 756 mM. and 18°

Found: 57,95% C; 4,18% H; 8,55% N.

Calculated for $C_8H_7NO_3$: 58,18% C; 4,24% H; 8,48% N.

The oxime crystallises from water in large white needles; m.p. 110°.

0,1242 gr. yielded 0,2418 gr. CO_2 and 0,0507 gr. H_2O

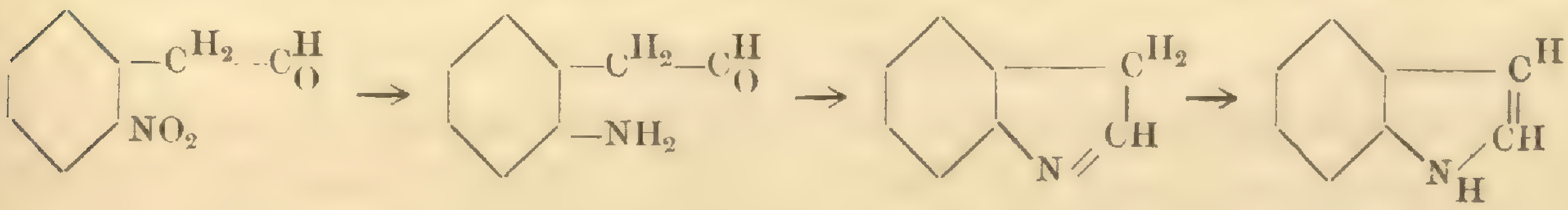
0,0907 gr. yielded 11,9 cc. N_2 at 775 mM. and 16°

Found: 53,10% C; 4,54% H; 15,53% N.

Calculated for $C_8H_8N_2O_3$: 53,33% C; 4,44% H; 15,55% N.

The aldehyde is characterised by the strong reddening of the alkaline aqueous solution. It further gives the ordinary reactions for aldehyde.

Reduced with iron powder in bisulphite solution it yields indole:



The idea of preparing the indole from orthonitrophenylacetaldehyde originated with A. VON BAEYER [1]); he did not, however, realise his object, as his efforts to prepare the aldehyde ended in failure; up to the present this substance has not been prepared.

The methyl *m*- and *p*-nitrostyrylaminoformates give in an analogous manner the corresponding aldehydes.

A method has thus been given to reduce the amides of the cinnamic acids to the aldehydes containing one carbon atom less. (I have not as yet tried the reaction with other $\alpha\beta$ unsaturated acids).

Indole.

For the practical preparation of the indole it is not necessary to prepare first the free aldehyde but the methyl orthonitrostyrylaminoformate is reduced in alcoholic solution with dilute acetic acid and iron powder, and the whole is then made strongly alkaline. This causes the urethane to saponify and the vinyl alcohol formed then condenses at once to indole which is then distilled in steam. The distillate is so charged with indole that this crystallises in the receiver. This indole is quite white and pure; m. p. 52°.

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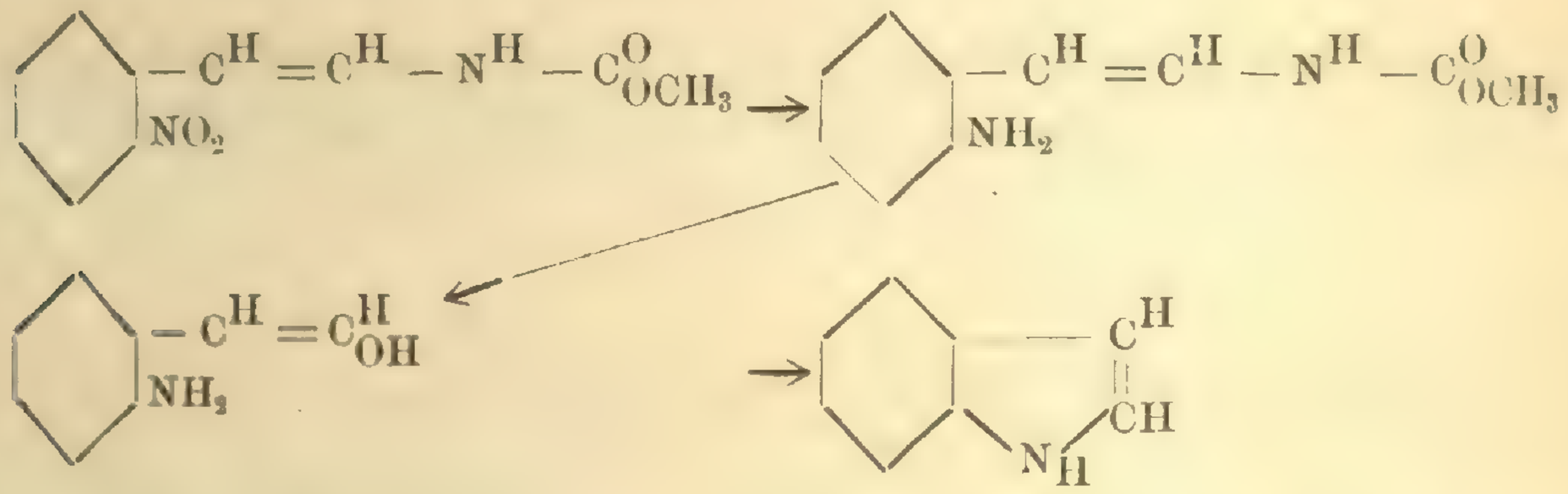
[1]) Lecture by A. v. BAEYER Ber. 33, Sonderheft blz. LVIII.

0,1533 gram yielded 16,2 cc. N_2 at 778 m.m. and 21°

Found: 12,26 °/₀ N

Calculated for C_8H_7N: 11.97 °/₀ N

With sodium nitrite and sulphuric acid it gives the well-known nitroso-indole reaction, and with picrit acid and sulphuric acid the picrate, which crystallises from benzene in red needles.



Delft, April 1909. *Chem. Lab. Techn. High School.*

Anatomy. — "*On the motor facialis- and abducens-nucleus of Lophius piscatorius.*" By Mr. A. B. Droogleever Fortuyn. (Communicated by Prof. L. Bolk). Communication from the Central Institute for Brain Research at Amsterdam.

(Communicated in the meeting of March 27, 1909).

When we study a series of transverse sections through the cerebrum of Lophius piscatorius, it is striking how here, in distinction from so many other fishes, the nucleus of the motor facialis lies partly dorsally, i. e. against the fourth ventricle, partly ventrally against the circumference of the medulla oblongata. Fig. 1 clearly shows this situation (d. fac. nu. and v. fac. nu.). For in other fishes, e. g. Gadus, we find the entire nucleus lying pretty near the ventricle, or we see, as in Tinca, that a few cells of this nucleus are displaced a little more ventrally, not so far, however, as to reach the circumference of the medulla oblongata. Fig. 2 is a transverse section of the most ventral part of the facialis-nucleus of Gadus, whilst Fig. 3 shows the facialis-nucleus of Tinca.

Now Dr. Ariëns Kappers, in a number of cases, has proved that such nuclear displacements, whether they come about in the course of the evolution, or, as with Lophius, are exceptional cases, always take place in the direction and under the influence of those tracks along which the most powerful and the greatest number of stimuli are conveyed to this nucleus. Kappers called this phenomenon of nuclear displacement neurobiotaxis.

Now, are there sufficient reasons to take for granted, that the removal of the motor facialis-nucleus in Lophius is likewise a case of neurobiotaxis?

In order to answer this question it should in the first place be settled towards which track the facialis-nucleus is shifted and next it should be explained why exactly this track in Lophius conducts stronger stimuli than in many other fishes.

Three tracks, which have a more or less ventral situation, may be considered to cause the shifting of the facialis-nucleus. These are the tractus tecto-bulbaris, (Fig. 1, tr. t.-b.), the radix descendens trigemini (Fig. 1, r. d. trig.) and the secondary gustatory tracks (sec. g. tr.) with the reticular substance surrounding them and the lobobulbar tracks. Dendrites of the motor facialis-cells have been traced in the areal of all these tracks.

Now apart from the fact that the shifting is more towards the tractus tecto-bulbaris than towards the two other tracks, still better arguments may be advanced why the secondary gustatory tracks and the radix descendens trigemini are not the cause.

Suppose the radix trigemini to draw the facialis-nucleus in a basal direction, then we should expect the trigeminus-nucleus itself to be still more strongly influenced. Now this is by no means the case in Lophius. And if we look at Gadus, an animal where, as appears also from Figs. 1 and 2, the radix descendens trigemini is about three times as thick as in Lophius, compared with the thickness of the oblongata, we see that there the facialis-nucleus is still partly situated close against the ventricle, partly shows a slight depression (Fig. 2), though not nearly so much as in Lophius.

Therefore it cannot be attributed to the radix descendens trigemini, that the facialis-nucleus in Lophius lies more ventrally.

The secondary gustatory tracks, which, owing to their position, might be the cause of the nuclear shifting, are but very slightly developed in Lophius. In Tinca, the tench, on the other hand, very large gustatory tracks are met with (Fig. 3, sec. g. tr.) and yet the facialis-nucleus, though situated not quite dorsally, is by no means so far displaced as in Lophius. The secondary gustatory tracks can therefore hardly be considered as the cause of the greater displacement in Lophius. The same holds good for the lobo-bulbar tracks because they are much smaller in Lophius than in Gadus and Tinca.

Preparations from Tinca strengthened the supposition that the tecto-bulbar track was the point of attraction for the facialis-nucleus as also here it was clearly to be seen that numerous protoplasm-offshoots of the ganglion-cells (dendrites) of the facialis-nucleus were

ramified amidst the fibres of the tecto-bulbar track. This phenomenon is also indicated in Fig. 3.

These tecto-bulbar tracks, which on the whole are very distinct in fishes, are especially in Lophius very large in comparison with the above-mentioned systems which are minute there, and a large part of these tracks, especially of the homolateral system, terminates in the region of the ventral facialis-nucleus.

It is a well-known fact that the tecto-bulbar tracks, originating in the terminal region of the optic nerve, transfer optical impressions on the motor systems of the medulla oblongata.

Now we may indeed take it for granted that the visual sense in Lophius is better developed than in many other fishes. If e. g. we compare the cerebrum of Lophius with that of Gadus, we see that the tectum opticum of Lophius is greater with respect to the other parts of the brain, telencephalon and cerebellum as well as the inferior lobes. The proportion of the tectum opticum with regard to the other parts of the brain is in Lophius about twice as large as in Gadus.

Moreover, as we saw above, taste is not much developed in Lophius, so that the eye appears to be its principal sense.

In order to determine how these optical reflexes can affect the facialis-system in Lophius, it was in the first place necessary to know which muscles were innervated by the motor facialis. They proved to be: the musculus adductor arcus palatini, the musc. hyomandibularis, the musc. adductor operculi, the musc. hyohyoideus superior, viz. the muscle which passes into the membrana branchiostega, and two muscles which are difficult to distinguish from each other, the musculi geniohyoideus et hyohyoideus inferior.

In most fishes the facialis also innervates the musc. levator operculi, but here, by way of exception, the vagus does so.

Since Lophius, as we know, has a comparatively huge head, and a body, which tapers immediately behind it and ends in a tail, all facial muscles are comparatively much larger than in other fishes.

This holds good particularly for the membrana branchiostega or the musc. hyohyoideus superior, which in Lophius appeared to be relatively about four times as large as in a fish with normal proportions, for which again Gadus was taken.

When the strongly developed facial muscles contract, they close the gill-cavity and help to open the mouth, and now the connection between visual impressions and contraction of the facial muscles becomes obvious.

For Lophius does not swim about, but generally lies half buried in the sand, so that only the dorsal part of its head and back

Fig. 1. *Lophius piscatorius.*

Transverse section of the medulla oblongata at the level of the facialis-nucleus.

IV = 4th ventricle.
M. v. = lateral vessel.
d. fac. nu = dorsal facialis-nucleus.
v. fac. nu = ventral facialis-nucleus.
tr. f. b. = fasciculus tectobulbaris.
r. d. trig. = radix descendens trigemini.
f. l. p. = fasciculus longitudinalis posterior.
mot. fac. = fasciculus motorius facialis.

Fig. 2. *Gadus morrhua.*

Transverse section of the medulla oblongata at the level of the most ventral part of the facialis-nucleus.

IV = 4th ventricle.
fac. nu = facialis-nucleus.
r. d. trig = radix descendens trigemini.
sec. g. fc = secondary gustatory tracts.
s. fac. = root of the sensory facialis.
mot. fac. = root of the motor facialis.
f. l. p. = fasciculus longitudinalis posterior.
tr. f. b. = fasciculus tectobulbaris.

Fig. 3. *Zeus faber.*

Transverse section through the medulla oblongata at the level of the facialis-nucleus.

IV = 4th ventricle.
fac. nu = facialis-nucleus.
tr. f. b. = tractus fasciculi bulbaris.
r. d. trig. = radix descendens trigemini.
sec. g. fc. = secondary gustatory tracks.
s. fac. = root of the sensory facialis.
f. l. p. = fasciculus longitudinalis posterior.

especially its angle and eyes appear above it. With this angle it attracts small fish and other animals, and when its prey has come sufficiently near, it jumps at it with a jerk of a fin or of its tail, and devours it. In doing this, however, it has to take care to keep its gill-cavities closed, these being so wide that a swallowed victim would be able to escape by this way.

Accordingly, whenever a prey comes near and gives strong visual stimuli, the facial muscles begin to work, because these stimuli are conducted to the facialis-nucleus along the tractus tecto-bulbaris [1]).

The shifting of the facialis-nucleus must therefore depend on the force and number of these stimuli, which other fishes do not nearly receive to the same extent.

That the sense of touch of the angle very likely plays no part in the catching of the prey, appeared from the slight development of the sensory nerves, leading to this organ.

In connection with the considerable size of the tectum opticum and the dorsal position of the eyes of Lophius (which in Gadus and Tinca have a lateral position) and their probable power of collaboration (while the field of vision in Gadus and Tinca is for the greater part, if not exclusively, unilaterally panoramic), it is also striking that part of the abducens-nucleus in Lophius, has already assumed a dorsal position, by the side of the coordinating system of the posterior longitudinal bundle. In the other above-named Teleosteans, though they are better swimmers, this nucleus lies still quite ventrally.

Summarizing I therefore think the following conclusions may be drawn:

1. In Lophius piscatorius part of the motor facialis-nucleus is displaced towards the tractus tecto-bulbaris, which conducts particularly powerful stimuli from the tectum opticum to the facialis nerve, because the sense of sight is well developed and in catching a prey, after stimulation of the eye, the gill-cavity must be closed and the mouth be opened, by a strong reaction of the particularly strongly developed facial muscles.

2. Contrary to other Teleosteans (Gadus, Tinca) part of the abducens-nucleus lies dorsally by the side of the posterior longitudinal bundle, a phenomenon which is more pronounced either in animals with a strongly developed and well-coordinated motility of the body or in animals with well-coordinated ocular movements.

3. The musculus levator operculi is here not innervated by the facialis-nerve, but by a branch of the vagus.

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[1]) It would be desirable to check all this in the living animal, but Lophius refuses to take any food in captivity.

Physiology. — *"On the consumption of oxygen by cold-blooded animals in connection with their size."* By Mr. F. J. J. BUYTENDIJK. (Communicated by Prof. T. PLACE).

(Communicated in the meeting of March 27, 1909).

The tables of the majority of investigators prove that in cold-blooded animals the consumption of O_2, as well as the production of CO_2 per kilogram of weight of the animal, is more considerable with small than with bigger animals of the same kind.

As an example I quote some figures from POTT [1]).

Rana temporaria	(old)	weight	13.9	gr.	CO_2	p.	K.G.	and	hour	180 cc.
,, ,,	(young)	,,	1.26	,,	,,	,,	,,	,,	,,	648 cc.
Bufo variabilis	(old)	,,	15	,,	,,	,,	,,	,,	,,	220 cc.
,, ,,	(young)	,,	3.6	,,	,,	,,	,,	,,	,,	770 cc.

KUNKEL [2]), on the other hand, in an accurate investigation of Limax variegatus, found proportionality with the volume.

The experiments with fishes of JOLYET and REGNARD [3]), BAUMERT [1]) and others nearly always proved the more intense respiration of the smaller animals.

Thus e.g. according to JOLYET and REGNARD the consumption is:

	Weight	O_2 p. K.G. hour	temp.
	Gr.	Cc.	
Cyprinus auratus	33.40	45.8	12°
,, ,,	82.130	35.03	12°
On the other hand by Muraena anguilla	51	40.5	14°
,, ,,	112	48	15°

More recent investigations of ZUNTZ (1901) [3]) with an apparatus constructed by himself lead him to the following conclusions, among others: "Die Grösse des auf die Gewichtseinheit bezogenen Verbrauches (von O_2) ist bei kleinen Fischen erheblich grösser und geht annähernd proportional der Körperoberfläche, ein weiterer Beweis dafür dass diese Beziehung nicht durch das Bedürfniss der Erhaltung der Eigenwärme zu erklären ist."

(The amount of oxygen consumed, referred to the unit of weight,

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[1]) According to ZUNTZ in HERRMANN's Handbuch der Physiologie Bd. IV, 2 S. 129—154.
[2]) KUNKEL, Verhandl. Deutsche Zöol. Gesellsch. Bd. 10 p. 22—31.
[3]) ZUNTZ, Verhandlungen der Berliner physiol. Gesellschaft XIII, 1901.

is with small fishes considerably greater and is approximately proportional to the surface of the body, another proof that this relation is not to be explained by the necessity of preserving the heat of the body).

These experiments, as far as I could gather from the short paper, were made with carp [1]). Also KNAUTHE [2]) experimented with this fish in ZUNTZ' laboratory. From his table it appears that the CO_2 production and the O_2 consumption of $2^1/_2$ year old carp is even somewhat greater than of $1^1/_2$ year old animals. His further experiments concerning the N-metabolism taught him "dass nicht die grössten sondern mittelgrosse Thiere den geringsten N-Umsatz zeigten."

("That not the largest, but medium-sized animals showed the greatest N-metabolism)."

VERNON [3]) determined the O_2 consumption in invertebrate sea-animals and also gives a table in which the weights of the experimental animals are mentioned. Some figures are repeated here.

	Weight	Average O_2 value at 16°
Rhizostoma pulmo	62.2	0.0167
	69.6	0.154
	87.1	0.083
	93.	0.106
	107.	0.060
Carmarina hastata	15.2	0.088
	27.6	0.065
	30·2	0.100
	30.7	0.129
	34.5	0.085
	44.8	0.088
	54.7	0.078

The rest of the table shows several deviations from a constant decrease of the O_2 consumption with the size, as is also the case with Carmarina. The author thinks that perhaps "the preservation of osmotic equilibrum between the tissues of the organism and the external medium may need the continuous application of vital energy and as the smaller the animal the larger the proportionate surface exposed a larger respiratory activity would necessarily result."

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[1]) The "Landwirthschaftlichen Jahrbücher", which were to publish more detailed accounts, I could not get at.

[2]) KNAUTHE. Arch. f. d. ges. Physiol. 73, S. 490—501.

[3]) VERNON. Journal of Physiology vol. 19 no. 1 1905.

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2. The method followed by WINKLER [1]) to determine O_2 in water is sufficiently accurate to follow the O_2 consumption of marine animals. In short it comes to this: whilst the air is entirely shut off a strongly alkaline JK solution and $MnCl_2$ is added to the liquid to be examined. The precipitate that is formed is, after settling, dissolved with HCl. The iodine that has been set free, titrated with $^1/_{100}$ norm. thiosulfate, yields with the aid of a simple formula (given by WINKLER) the quantity of O_2 per Litre.

That this method gives sufficiently accurate results, appears e. g from the following experiment:

Scyllium canicula:

Weight	temp.	Duration of the experiment	O_2 consumption	O_2 per hour and KG.	Difference
24 and 25 gr.	12.13°	1 hour	2,55 c.c.	52.04	
					0,78
id.	13°	2 hours	5,02 ,,	51,26	

In the following tables the O_2 consumption is calculated per hour and M^2. In order to convince myself, that the area of the animals may be said to be approximately proportional to the $\sqrt[3]{\text{weight}^2}$, I defined the area of some sharks by the following method.

The killed fish was dried in filter-paper and weighed. Then it was immersed in melted paraffine (50°), taken out and allowed to dry and cool, after which it was weighed anew. Now in different places of the animal bits of paraffine were taken off (pieces as flat as possible), so that the skin was laid bare. These pieces were weighed and measured (with millimetre-paper) and the average weight of 1 mm.^{2} of the layer of paraffine was determined. Thus I found:

Weight scyllium	Weight with paraffine	Paraffine	Weight 1 mm.^{2}. paraffine	Area in cm^2.	$\sqrt[3]{\text{weight}^2}$
9.9	11.65	1.65	0.5	33	4.64
108.270	114 850	6.580	0.36	183	22.7

With the small scyllium, therefore, $\sqrt[3]{\text{weight}^2}$ has to be multiplied by 8, with the large animals by 7, in order to get the area.

3. The experiments with Scyllium cassicula were made in a sufficient quantity of water in diffuse daylight, the sharks lying

[1]) WINKLER, Berichte d. Deutschen Chem. Ges. XXI, 2 p. 2843.

quiet on the bottom (not to mention an occasional movement) and breathing calmly. The duration of the experiments varied; the values of table I refer only to those cases when the animal after the experiment behaved quite normally. For in too long continued experiments asphyxiation sets in and the O_2 value becomes too small.

TABLE I.

Weight in grams	Cc O_2 per hour and individual	Cc O_2 per hour and K.G.	$\sqrt[3]{\text{weight}^2}$	Area	Cc O_2 per hour and M^2.
14	0 87	62.1	5.81	46.48	190.4
24	1.26	52.5	8.32	66.56	189.4
150	4.9	32.7	28.2	197.4	247.7
215	8.16	38	35.9	251.3	324.7
260	9.63	37	40.8	285.6	323.4

From the publications of MONTUORI [1]), who in different fishes stated the diminution of the O_2 consumption with the temperature of the water, I borrow the following scattered figures about Scyllium canicula.

Weight	Temp.	Cc O_2 per K.G. and hour	Temp.	Cc O_2 per K.G. and hour
200	14°	45	32°	22.88
39.5	14°	60	25°	40
11.8	12°	126.38	26°	51.61 or 74.77 [2])

If from these observations the consumption per M^2 is calculated for the same temperature, we find:

TABLE II.

Weight in grams	Temp.	$\sqrt[3]{\text{weight}^2}$	Area	Cc O_2 per hour individual	Cc O_2 per hour and M^2.
200	14°	34.2	239.4	9	375.9
39.5	14°	11.58	92.64	2.37	355.8
39.5	25°	11.58	92.65	1.58	170.6
11.8	26°	5.18	41.44	6.09 or [8.82]	147 [or 213]

[1]) MONTUORI. Zentralblatt für Physiologie Bd. XX n°. 8.
" Gazetta Internazionale de medicina Anno IX 1906.
[2]) Differs in the German and the Italian publication.

It seems to me that we are not entitled to conclude from these figures that the O_2 consumption is proportional to the surface.

Tables I and II only prove that the small Scyllium consume more O_2 than the big ones and that, calculated for the weight, this difference is most considerable with the smallest animals, without, however, reaching such a value as to become proportional to the area.

As to the cause of the greater intensity of respiration with smaller animals, it is in the first place the influence of the movements that should be considered. For these are quicker with smaller individuals (also with warm-blooded ones); moreover, in these experiments, the movement through the water will depend not only upon gravitation but also upon the friction at the surface.

The Scyllium, to be true, are during the investigation nearly always at rest. That also the tonus of the smaller animals is larger I venture to surmise a priori. In the second place the difference in age should be considered, which plays an important part also with mammals.

4. Probably very slowly growing animals, which during the experiments remain perfectly motionless, are the Scorpaena ustulata.

The experiments made with these had the following result:

TABLE III.

Weight in gr.	Cc O_2 p. hour individual	Cc O_2 p. hour and K.G.
8	0.38	47.5
21	0.9	45
55	2.5	45.5

With these animals as well as with the following I was unable to determine their surface. It appears, however, from Table III that the figures per K.G. and hour are about the same.

5. As to the influence of the movements on the O_2 consumption. I may say that a Crenilabrus occilatus, swimming regularly in broad daylight, consumed 0,56 Cc O_2 per hour, 0,365 in the dark, when the animal was not nearly so active.

I then compared two nearly full-grown individuals of Crenilabrus occilatus and Crenilabrus pavo in the dark.

TABLE IV.

		weight	O_2 per hour and individual	O_2 per hour and K.G.
Crenilabrus occilatus		10	0,365	36,5
,,	pavo	180	5,13	29,4

With a proportion of 1 : 18 in weight the O_2 consumption is in ratio of 1,24 : 1. With Scyllium a ratio of 1 : 18,5 agreed with a diminution of 1,7 : 1. Eliminating in the experiments with the Crenilabrus the influence of age, we could not help introducing other influences (movements, differences of species).

6. In connection with the above-mentioned experiments of VERNON I also investigated the O_2 consumption in a few individuals of Octopus vulgaris and of Echinus. Here it was my intention to compare two cold-blooded animals, one having a very active metabolism and a very rapid growth [1]), the other consuming very little O_2. In broad daylight the three Octopus individuals just caught, were quiet, their respiration was regular, after the experiment the animals were entirely normal. The smallest Octopus remained during the experimental period in a corner of the bottle and kept very quiet, the larger ones now and then put out a tentacle.

TABLE V.

	Weight in gr.	Cc O_2 per hour and individual	Cc O_2 per hour and K.G.
Octopus vulgaris	10	0.52	52
	110	3.01	27.36
	370	6.1	17
Echinus	45	0.106	2.3
	20	0.034	1.70

I may as well remark here that also Rhizostoma must have a very rapid growth, as animals of 2 or 3 K.G. may occur and the lease of life of these pelagic animals is not long. Carmarina, on the other hand, in which the difference in O_2 consumption is much less dependent on the size, not nearly reaches such a size (at most ± 100 gr.) and has not such a rapid growth.

The figures of my tables, like those of VERNON, show therefore

[1]) Dr. Lo BIANCO was so kind as to give me information about this.

in my opinion that the O_2 consumption of the small animals is considerably larger than that of the bigger ones of the same kind. This difference is, I believe, to be attributed to the influence of the more rapid motions of the smaller animals and the more intense metabolism which a growing body possesses. TIGERSTEDT [1]), therefore, rightly says that in general, where assimilatory processes take place, they are accompanied by active dissimilatory ones. Here also belongs without any doubt the observation of WARBURG [2]) that the O_2 consumption after fertilisation of the eggs of the sea-hedgehog increased by 6 and 7 times the amount. The action of light, osmotic pressure and liquid-pressure, which might cause a certain influence of the area on the respiration, has not been settled. The influence of light I have not been able to separate from variation of motility.

Naples, Febr. 8, 1909.

Microbiology. — *"The decomposition of uric acid by bacteria."* By Mr. F. LIEBERT. (Communicated by Prof. M. W. BEIJERINCK).

(Communicated in the meeting of April 23, 1909).

1. *Historical.*

The first investigators who studied the decomposition of uric acid by the action of bacteria were FAUSTO and LEONE SESTINI [3]).

They observed that uric acid in water when exposed to the air did not change, but that, as soon as the mixture was inoculated with a drop of rotting urine, the uric acid totally disappeared in some days.

Only when the experiment was interrupted before all the uric acid had been converted, they could detect urea.

Their conclusion was that the course of the process might be represented thus:

$$C_5 H_4 N_4 O_3 + 3\,O + 8\,H_2 O = 4\,NH_4 HCO_3 + CO_2$$

whereas as intermediate products alloxan and urea should occur, which latter substance they could really detect, but the former not. They have not worked at all with pure cultures.

The result obtained by E. GÉRARD [4]) was that two processes took place simultaneously.

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[1]) Handbuch der Phys. Nagel. Bd. 1 2e Hälfte 2e Heft.
[2]) WARBURG, Hoppe Seyler Zeitschr. f. Phys. Chem. Bd. 57 blz. 1.
[3]) Landwirtsch Versuchsst. 1890, Vol. XXXVIII, p. 157.
[4]) Comptes rendus T. 122, p. 187 et T. 123, p. 185.

First, the bacteria should decompose the uric acid into urea and tartronic acid; secondly, the urea should be hydrolised to ammonium carbonate by the urea bacteria. According to my own experiments I cannot accept the first part of this supposition, as the formation of tartronic acid is but an hypothesis, GÉRARD not having been able to find this substance. But the presence of urea amongst the products he could point out.

The first who studied the action of pure cultures on uric acid was C. ULPIANI [3]). He obtained a micrococcus which attacked uric acid easily, and found that the maximum and minimum temperatures for this organism were respectively 40° and 29°, and he presumed that its optimum temperature should be 39°. This latter statement is evidently wrong and his supposition, too, that the isolated bacterium cannot assimilate glucose, clearly reposes on an erroneous experiment.

At his request CINGOLANI ascertained that the equation of the process is:

$$C_5 H_4 N_4 O_3 + 2 H_2O + 3 O = 2 NH_2 CO NH_2 + 3CO_2.$$

The decomposition of uric acid by microbes has been studied by me after four methods. In the first place as an aërobic process; secondly as an anaërobic fermentation; thirdly as a denitrification; and finally as a sulphate reduction. As to the last I can be short: under none of the conditions chosen I could obtain with uric acid a sulphate reduction by bacteria.

As to the other cases, seven species of bacteria called for particular attention; the six first are aërobic bacteria, the seventh is a real anaërobic.

They are: 1. Bacillus fluorescens liquefaciens; 2. Bacillus fluorescens non liquefaciens; 3. Bacterium calco-aceticum (which is probably a new species but has already been obtained in different other ways in this Laboratory, where it is kept in culture by that name); 4. Bacillus pyocyaneus; 5. an aroma-producing species, which I wish to call *Bacterium odoratum*; 6. a urea-splitting bacterium *Urobacillus Musculi* and 7. a bacillus which causes an anaërobic fermentation in uric acid, *Bacillus acidi urici*.

The three last mentioned bacteria were formerly unknown, or at least not yet produced in pure culture.

2. *The chemical transformation of uric acid by the bacteria of the aërobic flora.*

It was observed that in all the examined species under aërobic action the uric acid is oxidised by the bacteria. As it was probable

[3]) Rendiconti d. Acad. dei Lincei 1903, T. 12, p. 236.

that the species here active would produce compounds which are also obtained chemically by alcaline oxidation, I have inquired whether by their action allantoin originates from uric acid, and my result was that B. pyocyaneus, B. fluorescens liquefaciens, B. fluorescens non liquefaciens and B. calco-aceticum do indeed oxidise uric acid after the equation

$$C_5 H_4 N_4 O_3 + O + H_2O = CO_2 + \underset{\text{Allantoin}}{C_4 H_6 N_4 O_3}$$

whereas by the same bacteria the allantoin is further oxidised and hydrolysed thus:

$$C_4 H_6 N_4 O_3 + H_2O + O_2 = 2\,NH_2\,CO\,NH_2 + 2\,CO_2,$$

whereby consequently urea is formed.

In nature also allantoin is met with. It was found in the urine of sucking calves, in the allantois liquid of cattle, and in the juice of Beta vulgaris. Man also secretes it after the use of food containing tannic substances.

The urea as well as the allantoin I could point out chemically as follows: Into a large 1 L. ERLENMEYER flask, 1 G. of uric acid was filled, 150 mG. of K_2HPO_4 and 300 cM^3. tap-water. After sterilisation this liquid was inoculated with the bacterium to be tested; then cultivated at 30°. After the course of a week the greater part of the uric acid had disappeared.

In order to isolate the allantoin, bariumhydroxid was added to precipitate the phosphates and the remaining uric acid, after which the superfluous baryta was removed by sulphuric acid. Now the liquid was evaporated to crystallisation and alcohol was added. This produced a white deposit which after re-crystallisation from water showed the characteristic crystals of allantoin. The thus obtained substance was further identified by the following reactions:

1. $AgNO_3$ and a trace of ammonia give a white deposit which is soluble in nitric acid and in an abundance of ammonia.
2. Mercurinitrate produces a white precipitate.
3. FEHLING's cupric test-liquid is reduced after long boiling.
4. The murexid reaction has a negative result.
5. At the determination of silver in the silver-salt was found 40,61 % (calculated 40,75 %).

The urea was identified as follows: The alcoholic filtrate was vapoured to dryness and re-crystallised. Nitric acid gives the characteristic crystals of ureanitrate [1]); the substance also displays the beautiful

[1]) H. BEHRENS' Anleitung zur mikrochemischen Analyse. Heft 4. p. 18.

reaction of cupri-ammonium cyanurate [1]); with nitrous acid nitrogen is produced.

It was also biologically demonstrated that the thus formed substance was urea. To this end the crystals were dissolved in broth mixed with a quantity of bacteria, then kept for some hours at 50°. At this temperature there is no growth but the urea is transformed into ammonium carbonate, which in this experiment resulted in great abundance.

Use can also be made of the iris phenomenon [2]) [3]) by adding some of the substance to yeastwater gelatin and making a plate upon which a strong urea bacterium is put. The NEWTON-rings, then occurring, prove that the added material contains urea.

As the media with uric acid in decomposition, are rendered strongly alkaline by the action of the urobacteria on the urea, the successive production of two distinct floras of bacteria is observed, one characteristic of the feebly acid or neutral medium, and one proper to the alkaline condition of the medium.

The allantoin produced at the oxidation of uric acid is itself an intermediate product, it being subject to further decomposition, which is in accordance with the fact that it is much more inconstant than uric acid.

This conversion is analogous to that of the last named substance, and so here also is a flora of a neutral and one of an alkaline stage; the neutral flora produces an oxidation and an hydrolysis.

If a hydrolytic agent acts chemically on allantoin, then, after the degree of intensity of the hydrolysis, either allanturic acid and urea or glyoxilic acid results, whereas two molecules of the latter are converted into the more constant glycolic acid and oxalic acid.

Of these decomposition products, oxalic acid is also easily found in the biological hydrolysis. It could be proved that all the four bacteria which form part of the "neutral flora" of uric acid, namely B. fluorescens liquefaciens, B. fluorescens non-liquefaciens, B. calco-aceticum and B. pyocyaneus, produce this substance. This acid forms a deposit of calcium oxalate at the scratches of the vessel-walls. That the substance was indeed calcium oxalate was confirmed, besides by the characteristic forms of the crystals, by its solubility in hydrochloric acid, its insolubility in acetic acid and its power of decolorising acid permanganate. Moreover, after incineration a mixture of calciumoxid and calcium carbonate remained.

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[1]) See footnote 1 on the preceeding page.

[2]) BEIJERINCK, Centralbl. f. Bakter. Bd. VII 1901.

[3]) SÖHNGEN, Kon. Akad. v. Wetensch. 12 Nov. 1908.

The chemical oxidation of allantoin may give rise to several products, and finally calcium carbonate is formed.

In the biological oxidation of allantoin calcium oxalate also results. As it is probable that glycolic acid is at first produced it may be mentioned that in cultures, containing nothing but calcium glycolate as carbon food, which is not apt chemically to be converted into calcium oxalate, yet oxalate is produced by the bacteria. The process of oxidation does not however stop here, but the oxalate is further oxidised into carbonate; this takes place abundantly when to the liquid 0.05% calciumchlorid is added. The same occurs not only in accumulations with allantoin or calcium glycolate, but also with calcium oxalate as only carbon food, which is oxidised by the said bacteria, and chiefly by B. calco-aceticum; consequently this species is easily accumulated from garden soil with the said substance.

In order to control the rapidity with which the uric acid disappears in the pure cultures, the several isolated bacteria were put in tapwater to which a known quantity of uric acid and 0.1% bikalium phosphate had been added. Use was made of MERCK's acidum uricum pur. which was snow-white and contained no ponderable quantity of ashes. The acid was before weighing dried at 110° to constant weight. The rapidity of the decomposition may be determined in two ways; by controlling how much uric acid is converted in unit of time, and by determining the time in which the uric acid has disappeared. It seems that the former method is to be preferred here, as by the latter the retarding influence of the products of decomposition will be strongly felt and the results become less clear.

Nor will in the first mentioned case the determination of the decomposed uric acid offer any difficulty. After two days' growth at 30° the cultures were slightly acidified with strong hydrochloric acid and filtered through a GOOCH crucible. For washing was used a saturated solution of uric acid in water. In order to form a judgment of the error made in this way, 501 mG. of uric acid was added to a culture containing an abundance of B. fluorescens non-liquefaciens; analysing at the same time 499 mG. were found back, so the error is less than $^1/_2$ %.

If uric acid was used as sole carbon and nitrogen food, B. fluorescens liquefaciens assimilated 82 mG. in two days. For B. fluorescens non-liquefaciens, and B. calco-aceticum the quantities were respectively 85 and 96 mG.; the volume of the medium was 150 cM.^{3} and $^1/_{20}$ % K_2HPO_4 had been added.

Furthermore it was inquired whether the uric acid would be still

decomposed if at the same time other nutriment was present. To this end either $^1/_2$ % mannite or $^1/_2$ % peptone was added to the above culture liquid, which contained excess of uric acid. After two days mannite was still present.

	Uric acid assimilated in two days	
	in the solution with peptone	in the solution with mannite
B. pyocyaneus	151 mG.	91 mG.
B. fluorescens liquefaciens	82 "	48 "
B. fluorescens non-liquefaciens	41 "	35 "
B. calco-aceticum	79 "	60 "

These data prove that even if at the same time an easily assimilable substance is present, the uric acid is still attacked and is a good nutriment for the said species.

3. *Bacteria active in the aërobic decomposition of uric acid.*

As said above these bacteria belong to two consecutive floras; the first is found as long as the medium reacts neutrally or is feebly acid, the second, characteristic of the alkaline phase, may be called "urea-flora", as the urea, produced by the first flora, is by it converted into ammonium carbonate.

a. Flora of the acid or neutral phase.

Like my predecessors I used uric acid at the same time as source of carbon and of nitrogen. Two different liquid culture media were prepared:

1. The liquid of GÉRARD (l.c.): 1 G. of uric acid, 12 G. K^2HPO^4, 1000 cM.^{3} tapwater.

2. 1 G. of uric acid, 0.1 G. K^2HPO^4, 1000 cM.^{3} tapwater.

Between these two culture liquids there exists, besides in the rate of phosphate, a remarkable difference: In GÉRARD's liquid all the uric acid is dissolved and the reaction is neutral. In the other it is acid to the end, although feebly, by the excess of uric acid, even if by the bacteria ammonia is produced. Both liquids were filled into small ERLENMEYER flasks so that the layer was $\pm$ 1 cM. high. Garden soil was used for the inoculation. At 30° in both tubes a rich culture was observed after 2×24 hours, and the transfers also went very well.

The reaction of GÉRARD's liquid was then still neutral, whereas the other under these conditions reacted still feebly acid.

The flora which first appeared proved in both cases to be the same and consisted of B. fluorescens liquefaciens, B. fluorescens non-liquefaciens and B. calco-aceticum. After repeated transports B. fluor. liq. vanished completely, the optimum temperature of this species being nearer to 25°, whereas that of the other is about 30°. In the neutral liquid of GÉRARD this microbe could however maintain itself longer than in the other.

The isolation of the bacteria was done on broth-gelatine.

On agar-plates, which besides a little phosphate contained some uric acid, by which the whole plate was quite opaque, the uric acid disappeared around the bacterial colonies under the influence of the formed ammonium carbonate. As this compound increased a ring of crystals occurred in which calcium urate could be pointed out. If the growth took place not at 30° but at 37°, it was found that (from garden soil as well as from canal water) the same bacteria as from the cultures at 30° were obtained, with the exception of those cases when B. pyocyaneus occurred in the material, as this species dispels the others more or less completely.

Uric acid thus proved to be a very good food for this latter microbe too, although the power of secreting pyocyanin was lost and could only be restored by repeated transplantations in peptone solutions.

By many experiments it was proved that during the winter of 1907—1908, as well from canal water as from garden soil at Delft, by this method a culture was constantly obtained in which B. pyocyaneus positively predominated. But in 1909 no more a growth was to be had containing this species, and when material was used from a garden at the Helder, the results were also negative.

With a view to get more certainty about this point, I added to a rough accumulation with uric acid at 37° a trace of a culture of B. pyocyaneus. How small soever the number of introduced germs might be, they were always able to maintain themselves, even in the transports. It seems thus proved that the occurrence of B. pyocyaneus in the soil is but accidental.

b. Flora of the alkaline phase in the decomposition of the uric acid.

As has been described in the preceding pages B. fluorescens produces urea from uric acid. If a crude accumulation is allowed to stand it may thus be expected that the urea bacteria which, like B. fluorescens, were present in the original inoculation material, will find in the liquid containing urea the conditions wanted for their growth. This is indeed the case as is shown by the intense production

of ammonium carbonate, for a culture obtained in this way had a titre of 0,18 N. The urea flora did not however directly follow that of B. fluorescens and allied species, for when a rough culture is only some days old it evolves a pleasant odour of a compound ester, and cultures show that the original species are replaced by an aroma producing microbe. Milk used as medium for this bacterium favours very much the production of the aroma which after some days reminds of the flavour of young cheese. This bacterium, B. odoratum, does not however produce urease, although it is capable of splitting urea by catabolism.

To obtain the urea flora which succeeds that of B. odoratum, the culture was streaked on broth gelatin with addition of 1% urea and 0,1 % ammonium carbonate. But no ureabacteria developed, which did take place on yeastwater gelatin with the above additions. A description of the thus isolated single species was not found; it shall be called *Urobacillus Musculi*. The temperature of 30° is the most favourable for its culture as, especially on broth-agar urea and ammonium corbonate, then an abundance of urease-producing colonies occur. Sporulation has not been observed and gelatin is not liquefied. The maximum quantity of urea converted into a 1% peptone solution amounts to 3,6 % [1]).

Addition of peptone is not quite necessary for the splitting of urea, but it highly favours it [2]). Calcium malate, calcium lactate and glucose, together with urea, gave only rise to a very slight decomposition.

3. *Denitrification with uric acid as carbon food.*

If a stoppered bottle, quite filled with water and uric acid, with addition of 1 % kaliumnitrate and a little phosphate, is inoculated with soil, then after some days' cultivation at 30° under these anaërobic conditions, gas evolves, which proves to consist of a mixture of carbonic acid and nitrogen.

A plate culture on broth gelatin shows that the denitrifying bacterium, B. Stutzeri [3]), is cause of the process, hence this species may be very well accumulated in the said way. It also denitrifies strongly with uric acid in pure culture. In three days, for instance, 250 mG. KNO_3 were completely decomposed in 250 cM^3. of the culture medium, so that sulphuric acid and diphenylamin did no more react.

At denitrification in general the escaping nitrogen originates from the

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[1]) Beijerinck, Centr.bl. f. Bacteriol. II, VII Bd. 1901.
[2]) Söhngen, Kon. Akad. v. Wetensch 12 N 1908.
[3]) van Iterson, Centralbl. f. Bakteriologie II, XII Bd. 1902.

nitrate or the nitrite, and not from the concerned organic compound. With uric acid there was reason to suppose that this could be otherwise, as perhaps the amido groups might react in the feebly acid medium with the nitrous acid and thus per molecule of nitrate two molecules of nitrogen should escape. Quantitative tests have however shown that this was neither the case here.

The same denitrification experiment being repeated at 37°, B. pyocyaneus was obtained in the winter of 1907—1908. When this bacterium was not present in the soil, as in 1909, the denitrification remained feeble, 37° being too high for B. Stuzeri, which is then also the originator of the gas.

4. *Fermentation of uric acid by an anaërobic.*

In the same way as in the preceding case, stoppered, 100 cM.³ bottles were used for these experiments; they were filled with 1 G. of uric acid and further with tapwater to which 0,1 % kaliumphosphate had been added.

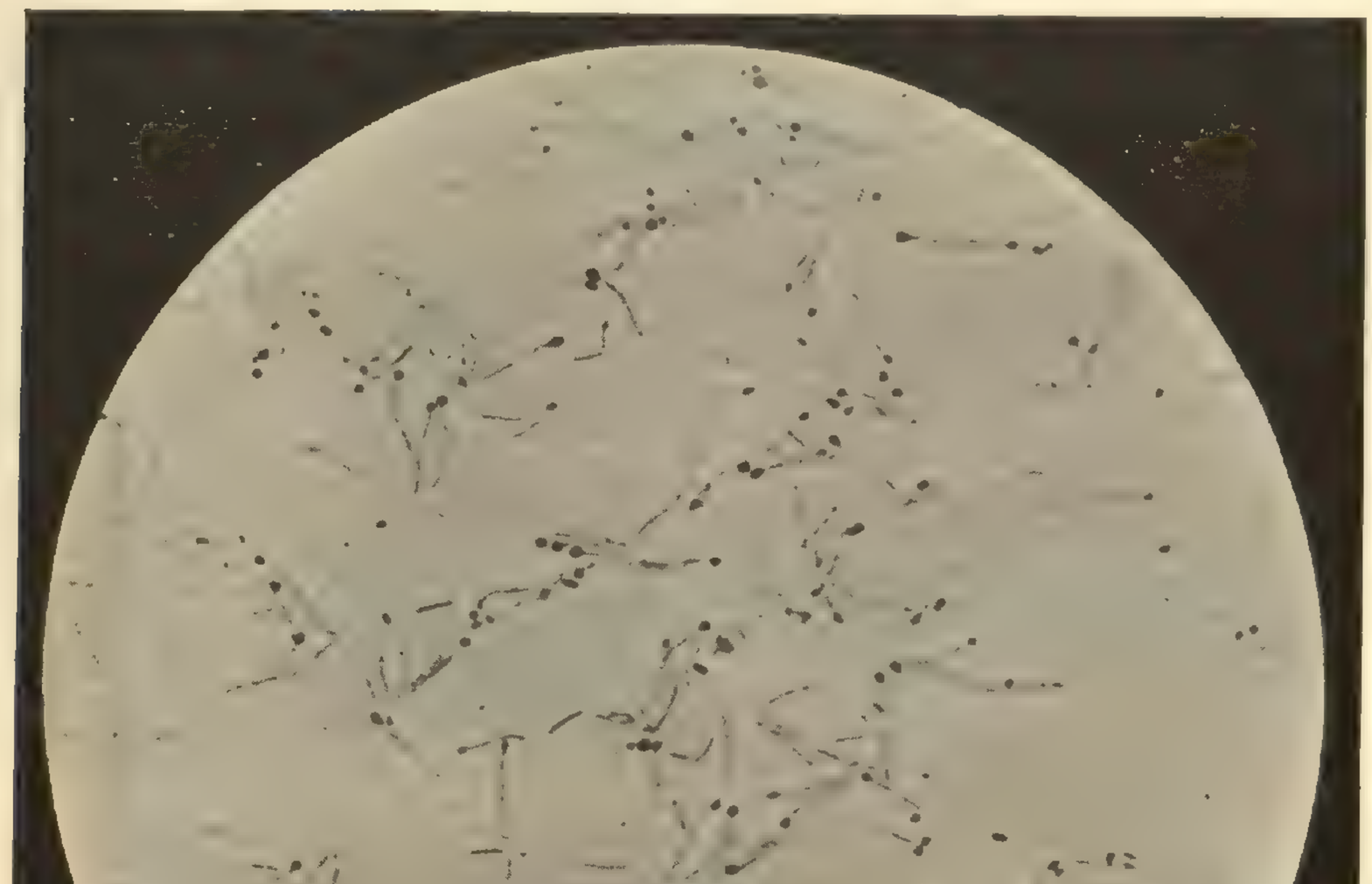
For the inoculation I took garden soil containing carbonates, from which by the free uric acid some carbonic acid evolves. This slight gas production however soon ceases. Canal mud, too, served well as inoculation material. With heath soil adhering to Erica roots, the process was slower and a greater quantity of inoculation material was wanted to bring about a good fermentation.

The most favourable temperature lies at about 35°; at 45° the fermentation is less energetic. Temperatures below 30° are also less fit, as then the fermentation does not always go on regularly.

After a day's cultivation the first traces of gas are already observed, and then it rapidly increases in quantity. But when it has reached a certain height the fermentation strongly relaxes. This relaxing could not be ascribed to want of uric acid, as by the murexidreaction it could yet in abundance be indicated, both in the fluid and in the precipitate.

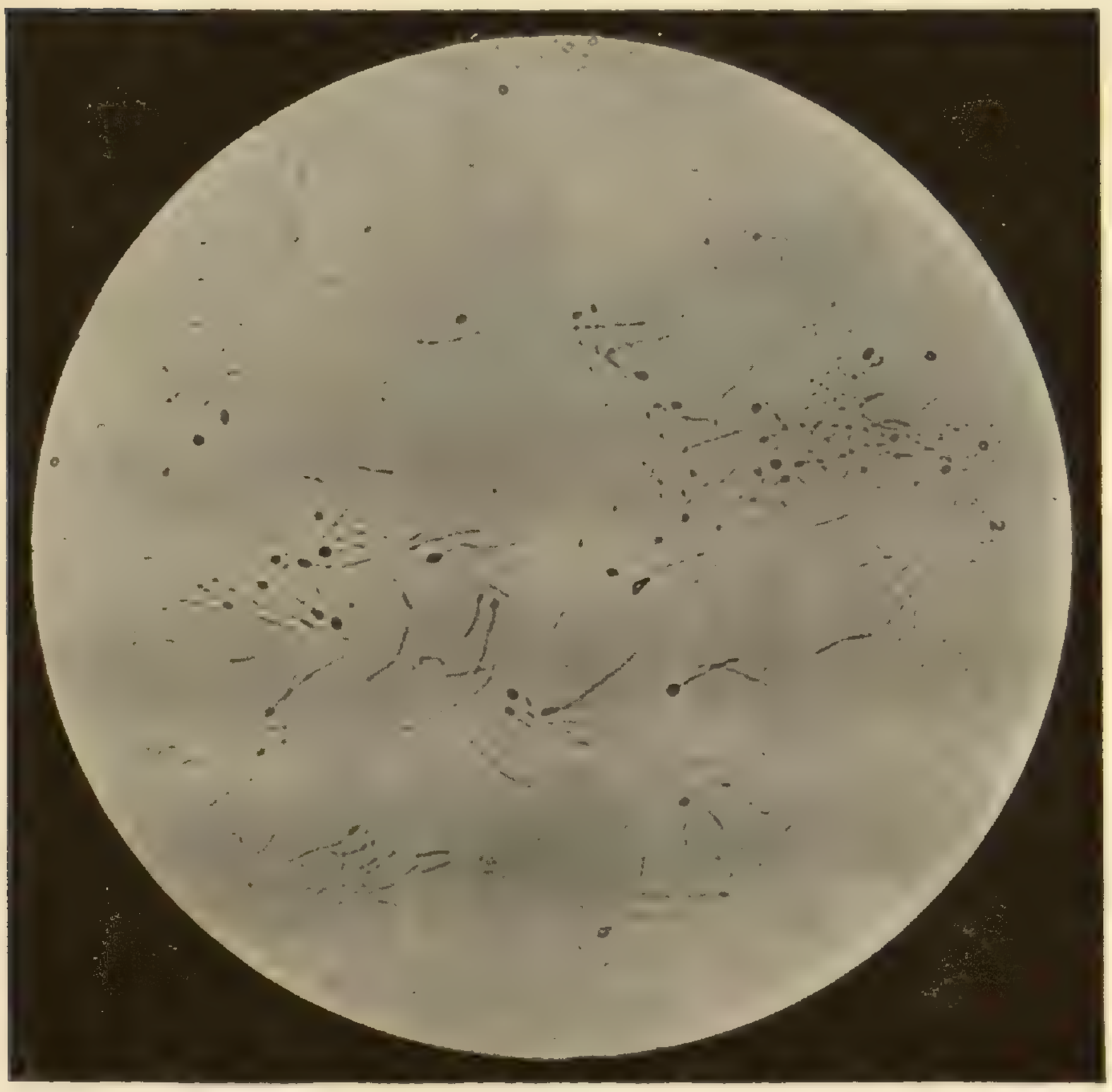
A new supply of uric acid did somewhat accelerate the fermentation, but as was to be expected, gave no positive amelioration. The great concentration of the ammonium compounds from the uric acid was found to be cause of the abatement. This noxious influence was however eliminated by the addition of neutral magnesium phosphate ($MgH\ PO_4 . 7\ H_2O$). This salt, with difficulty soluble in water (0.01 gr. per L.), combines with the ammonium carbonate to ammonium-magnesium phosphate,hence the reaction continues neutral. That the addition of magnesium phosphate really has this result is at once

Fig 1.



Bacillus acidi urici, coloured with carbol-fuchsine (720).

Fig 2.



Bacillus acidi urici, not coloured (720).

evident when comparing the image of a culture in its first phase of fermentation to that of one where the fermentation has ceased. In the former case the irregular uric acid crystals and the boat-shaped ones of the magnesium phosphate are only to be seen; in the latter the characteristic "coffins" of ammonium-magnesium phosphate.

In the deposit at the bottom we observe numerous highly motile rods, whose motility rapidly decreases at access of air; so, there is reason to believe that the excess of magnesium phosphate acts also beneficially on the bacteria, by enabling them easily to reach the places with the most favourable oxygen tension.

If the fermenting fluid is poured on the plate with admission of air, no microbes occur which ferment the uric acid; but B. calco-aceticum does develop.

The fermentation organism proper which was hitherto unknown, and shall be called *Bacillus acidi urici*, is a spore-forming, strongly motile, obligative anaërobic microbe, whose photographic image is given here.

The middle length of the rods is $5\,\mu$; their width is $0.7\,\mu$, and the diameter of the spores is about $2.5\,\mu$. The rods are excessively motile but by cultivation on plates the motility gets quite lost. The spores are round and placed at the ends, but eventually they are oval. The colonies on broth-agar are transparent, round or of more irregular shape, and very variable in size.

As to the products of fermentation the following was established. The escaping gas consists solely of carbonic acid. Quantitative tests proved that per one molecule of uric acid three molecules of carbonic acid are liberated. It was further observed that the urea groups are converted into ammonium carbonate, and that acetic acid is formed.

The quantity of this acid greatly depends on the nature of the fermentation; a crude accumulation produces more acetic acid than a fermentation that has been repeatedly transferred, or has been obtained by pure culture.

As to the chemism of the fermentation the production of carbonic acid corresponds with the formula as given by STRECKER [1]) for the hydrolytic decomposition of uric acid:

$$C_5H_4N_4O_3 + 5\,H_2O = C_2H_5NO_2 + 3\,CO_2 + 3\,NH_3$$

Glycocoll

But glycocoll I have not been able to point out. It is however very well possible that this substance is reduced to ammonium acetate by the further action of the bacteria.

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[1]) LIEBIG's Annalen: Bd. 146, pag. 142, 1868. Zeitschr. f. Chemie 1868 pag. 215.

It is also possible that the decomposition takes place after the equation:

$$C_5H_4N_4O_3 + 6\ H_2O = 3\ CO_2 + \underset{\text{Glycolic acid}}{CH_2OHCOOH} + 4\ NH_3$$

However I have as little been able with certainty to detect this acid [1]), but then, here too, the possibility exists that it is reduced to acetic acid through the influence of the bacteria, which point will be a subject for later inquiries.

Summary.

In the preceding it has been proved:

1. That when uric acid is attacked by aërobic microbes this acid is transformed into carbonic acid and ammonia, whereas as intermediate products allantoin, urea and oxalic acid are formed. The bacteria causing this chemism may be divided into two groups: a flora of the feebly acid or neutral phase of the medium, to which belong B. fluorescens liquefaciens, B. fluorescens non liquefaciens and B. calco-aceticum; and a flora which develops in the culture liquid after it has become alkaline, and which consists of *B. odoratum* and *B. Musculi*.

2. That B. pyocyaneus and B. Stutzeri with uric acid as carbon food, in presence of saltpeter, cause denitrification.

3. That under absolutely anaërobic conditions uric acid may be fermented by the spore-forming *Bacillus acidi urici* n. sp., in which process ammonia, acetic acid, and 3 molecules of carbonic acid per molecule of decomposed uric acid are produced.

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EXPLANATION OF THE FIGURES.

Fig. 1. *Bacillus acidi urici.* Magn. 720. (Zeiss. 2,5 mM. Proj. oc. 2). Colony on broth-agar, coloured with carbol fuchsine.

Fig. 2. *Bacillus acidi urici.* Magn. 720. Colony on broth-agar, taken after life.

[1]) In order to find this acid I made the fermentation take place in a Chamberland-bougie, quite immersed in sterile water, so that the intermediate products might diffuse in it; but it was of no avail.

Botany. — *"Investigations on the relation between the presentation time and intensity of stimulus in geotropic curvatures."* By Miss C. J. PEKELHARING. (Communicated by Prof. F. A. F. C. WENT).

The investigations of BLAAUW [1]) and of FRÖSCHEL [2]) have shown that in the case of phototropic stimulation by light of various intensities, the presentation time is inversely proportional to the intensity of the light.

It is not known whether a similar connexion exists for other kinds of stimuli; for geotropic curvatures, however, BACH's observations [3]) seem to indicate that this is indeed the case.

BACH examined the influence of various external conditions on the presentation time for geotropism. Some of his observations are concerned with the inertia forces resulting from small and large centripetal accelerations [4]), and with the influence of the angle at which the plant is fixed. As was shown by FITTING's experiments [5]), the plant is subjected in the latter case to a force equal to $mg \times$ the sine of the angle which the plant makes with the normal.

BACH himself does not deduce from his figures any relationship between the duration of the stimulus and its intensity. If such a deduction be attempted, and (for the sake of agreement with BLAAUW's method) the presentation time be expressed in seconds — if furthermore a mean is taken for the centrifugal forces which are given for each experiment, then the following table results:

I. With centrifugal force.

Acceleration	Presentation time	Product in g-seconds
1.07 g	480″	513
1.75 g	270″	472
3.00 g	180″	540
5.25 g	120″	630
10.65 g	60″	639
19.40 g	30″	582
27.35 g	15″	410
0.71 g	600″	426
0.60 g	1500″	900
0.40 g	1800″	720
0.14 g	3000″	420

[1]) A. H. BLAAUW, *a*. Proceedings of the Roy. Acad. of Sciences, Amst. Sept. '08. *b*. Die Perception des Lichtes. Recueil des Travaux Botaniques Néerlandais. Volume V. 1909.

[2]) P. FRÖSCHEL. Sitzungsberichte d. K. Akad. der Wiss., Wien, 117. 1908.

[3]) H. BACH. Jahrb. f. Wiss. Bot. XLV. 1907, p. 86—91.

[4]) I will call these centrifugal forces.

[5]) H. FITTING. Jahrb. f. Wiss. Bot. XLI. 1905, p. 282.

II. With positions at an angle.

Corresponding acceleration	Presentation time	Product in g-seconds
$g \times \sin 90° = g \times 1$	450″	450
$g \times \sin 60° = 0.866\, g$	600″	520
$g \times \sin 45° = 0.7071\, g$	690″	488
$g \times \sin 30° = 0.5\, g$	840″	420
$g \times \sin 15° = 0.259\, g$	1080″	280

FRÖSCHEL [1]) considers that these figures prove the existence of the relationship in question; the number of values is however small, the variations are very large and are for small accelerations of such a nature that a more extensive and accurate investigation was considered desirable by Prof. WENT [2]).

As BACH himself found, the presentation time for geotropic curvature depends very much on external conditions. The effective force cannot be varied within very wide limits (as in the case of light, where for instance a change from 1 to 140 million is possible). It is therefore necessary to have a large number of experiments, and to make the experimental error small; these requirements need not be satisfied in the investigation of phototropic stimuli.

I have therefore partially repeated BACH's investigation in Prof. WENT's laboratory. As far as possible I took precautions to keep the external conditions constant. The experiments with plants placed at an angle were made in a perfectly dark space in the laboratory glass-house, in which the temperature was kept pretty constant at 24° C. In order to exclude phototropic curvatures, the observations were made with a red electric incandescent lamp. In order to eliminate individual differences, each experiment was done simultaneously with at least 12 plants. I mostly used etiolated coleoptiles of *Avena sativa*; before the beginning of each experiment very long and very short individuals were removed from the experimental box. The seedlings had germinated in the same glass-house in which they were used, so that the influence of changed conditions at the beginning of the experiment, observed by BACH, was also excluded.

As a preliminary communication I here give a synopsis of the results of my experiments.

The following table gives for each angle a figure, indicating the period of stimulation, after which about half the seedlings had become curved.

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[1]) P. FRÖSCHEL. *a*). l.c. *b*). Oesterr. bot. Zeit. Jahrg. 1909, no. 2.

[2]) F. A. F. C. WENT. Proc. Roy. Acad. of Sciences. Amsterdam. Sept. 1908.

Corresponding acceleration	Presentation time	Product in g-seconds
$g \times \sin 90° = g \times 1$	269″	269
$g \times \sin 60° = g \times 0.866$	326″	282
$g \times \sin 120° = g \times 0.866$	332″	288
$g \times \sin 45° = g \times 0.7071$	366″	259
$g \times \sin 135° = g \times 0.7071$	340″	240
$g \times \sin 40° = g \times 0.6428$	441″	284
$g \times \sin 30° = g \times 0.5$	540″	270
$g \times \sin 150° = g \times 0.5$	538″	269
$g \times \sin 25° = g \times 0.423$	607″	257
$g \times \sin 20° = g \times 0.342$	735″	251
$g \times \sin 159° = g \times 0.358$	730″	262
$g \times \sin 15° = g \times 0.259$	871″	226
$g \times \sin 165° = g \times 0.259$	853″	220
$g \times \sin 10° = g \times 0.174$	1405″	244
$g \times \sin 171° = g \times 0.156$	1498″	234
$g \times \sin 5° = g \times 0.087$	2629″	229
$g \times \sin 174° = g \times 0.105$	2233″	234

In the case of the smaller angles with the normal (15° etc.), the deviation from the standard product (for $90° = 269$) is indeed fairly large but this is perhaps explained by the rapid change in the sine of these angles. The boxes in which the plants were grown could indeed be accurately placed in the desired position, but we cannot assume that all coleoptiles really grow quite vertically, and in these positions a deviation of $\cdot {}^1/_2{}^\circ$ produces such a change in the effective force, that great accuracy is thereby excluded.

The experiments with centrifugal force were carried out in the laboratory in a dark well-ventilated room in which the gas supply was permanently turned off, and at a fairly constant temperature of 17°. The presentation time for 90° is at that temperature greater than at 24°. This influence of the temperature had already been observed by BACH. I have found the same influence.

Thus I found for instance at a deviation of 90° a presentation time of 4′13″ at 26° C. and of 5′7″ at 21.5° C.; at a deviation of 60° 5′25″ at 24° C., 5′41″ at 21.5° C.; at a deviation of 30° 9′ at 24° C., 9′20″ at 22°.5 C.

The standard product is here 300. The seedlings were grown in annular boxes of various radii (up to 95 m.m.). The plants did not come up in a perfectly regular circle round the centre; as radius the mean had to be taken of distances, which differed amongst themselves by 2—4 m.m. This causes in the product an error of 3%. With the aid of Mr. D. B. KAGENAAR, scientific instrument maker

at Utrecht, an apparatus was constructed for these experiments which runs perfectly regularly, which gives accelerations from 0.03 g. to 60 g., which picks up and looses the whole of its speed in the space of $1^1/_2$ seconds, and which can be used with a vertical and with a horizontal axis.

With this apparatus I obtained the following results:

Acceleration	Presentation time	Product in g-seconds	Position of axis
0.08 g	3900″	312	horizontal
0.08 g	3900″	312	vertical
0.14 g	2230″	312	horizontal
0.25 g	1300″	325	,,
0.36 g	830″	299	vertical
0.38 g	805″	306	horizontal
0.60 g	510″	306	vertical
0.67 g	441″	296	horizontal
0.76 g	415″	315	,,
1.04 g	310″	322	vertical
1.25 g.	248″	310	,,
2.08 g	140″	291	,,
2.14 g	145″	310	,,
2.24 g	125″	281	,,
2.30 g	135″	311	horizontal
2.89 g	110″	318	vertical
3.00 g	100″	300	,,
3.04 g	95″	286	,,
3.71 g	80″	297	horizontal
3.93 g	75″	295	vertical
4.42 g	72″	318	,,
4.43 g	70″	310	horizontal
4.68 g	65″	304	vertical
5.36 g	55″	295	,,
5.76 g	53″	305	horizontal
6.48 g	45″	292	vertical
10.08 g	31″	312	,,
11.70 g	26″	304	,,
13.89 g	22″	306	horizontal
17.28 g	18″	311	vertical
23.86 g	13″	310	,,
41.76 g	7″	292	,,
58.43 g	5″	292	,,

In these products there is therefore a possible error of 3 % in

consequence of the nature of the experiment. The results for a vertical and for a horizontal axis are in agreement. For forces with small accelerations (0.12 g. and below) the intensity of the curvature becomes smaller and smaller, and finally (below 0.8 g.) it is difficult to judge. This perhaps explains a difference between BACH's results and my own for forces with accelerations below 0.7 g. In those cases BACH finds presentation times, which are relatively too high. He however also finds the reaction times longer than I do in the corresponding cases. For instance BACH found (with *Vicia Faba*) for 0.3 g. a reaction time of $1^h.55'$, i. e. 59 % longer than the reaction time for 1 g. I found myself (with *Avena sativa*) for 0.28 g. a reaction time of 50′ i. e. 10 % longer than the reaction time for 1 g. Similarly BACH: for 0.06 g. a reaction time which is 188 % longer than that for 1 g.; I for 0.06 g. a reaction time which is 147 % longer.

The roots of *Vicia Faba* are certainly a less sensitive material than the coleoptiles of *Avena*. Perhaps BACH stimulated in these cases for a somewhat longer time than the presentation time, in order to obtain curvatures which could be observed very clearly. The experiments are especially in this part of his investigation not sufficiently numerous to be convincing.

His attempt to explain the values which to him also appeared abnormally high, is certainly not justified, since it assumes, without any experimental basis, that there is a difference in the perception of the stimulus depending on whether the centrifuge has a vertical or a horizontal axis. In my own experiments I have not observed any such difference as will be seen from the above figures.

It follows from all these figures for centrifugal forces and for various inclinations, that the relationship, which it was thought could be deduced from BACH's experiments, does indeed exist.

I have also carried out some experiments with roots, namely with roots of *Lepidium sativum*, which were grown according to BUDER's method [1]). I obtained with these the following results:

Corresponding Acceleration	Presentation time	Product in g-seconds
$g \times sin\ 90° = g \times 1$	355″	355
$g \times sin\ 45° = g \times 0.7071$	544″	385
$g \times sin\ 135° = g \times 0.7071$	525″	371
$g \times sin\ 30° = g \times 0.5$	715″	358
$g \times sin\ 150° = g \times 0.5$	700″	350
$g \times sin\ 15° = g \times 0.259$	1460″	378
$g \times sin\ 165° = g \times 0.259$	1370″	355

[1]) J. BUDER, Ber. d. Deutschen Bot. Gesellsch. XXVI. 1908, p. 164.

The result of these experiments is therefore that, as in the case of phototropic curvatures, so also for geotropic curvatures the product of duration of stimulus and intensity of stimulus is constant. To what extent the stimuli of light and of gravity are comparable in their action on the plant, cannot be determined by these experiments alone.

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Geology. — *"The Fens of the Indian Archipelago."* By Prof. A. WICHMANN.

In 1778 FRANZ VON BEROLDINGEN propounded the theory, that lignite and coal measures are to be regarded as bog strata, of which the peat, after it became covered with other soils, was transformed into coal. This theory has so far triumphantly maintained itself. Nor is there any divergence of opinion concerning a second proposition, viz. that during the carboniferous period a uniform tropical climate was maintained over the whole of the globe, as is deduced by palaeophytologists from fossil vegetable remains. In disagreement with the above-mentioned theory, it is asserted in most text-books and in other publications, that peat formations, at least autochthonous ones, are not found in tropical countries. In order to escape from this contradiction some geologists assume that coal measures cannot be derived from fens, while others seek a solution of the problem by supposing that formation of coal, like that of peat, can only take place in a cool climate.

Although the presence of fens in the tropics has been pointed out repeatedly, and as far as the Indian Archipelago is concerned, JOHN ANDERSSON in 1794 mentioned the peat of Sumatra, these facts have been denied the attention which they really deserve. On the contrary, it has gradually become a sort of axiom, that fens cannot form a part of tropical countries. The many geographical works on the Indian Archipelago are for the greater part silent on the occurrence of fens, although they might have been better informed.

That such formations receive only scant attention, must be attributed to the fact that the vast majority of explorers, and also those who are called to occupy themselves with the investigation of the soil, possess a wholly inadequate knowledge of the subject. Moreover peat is of no economical importance to tropical countries, because the latter possess more than enough fire wood for domestic needs and because it is of no use for industrial purposes on account of its high

ash content. In contradistinction to the peat-bogs of temperate and cold regions, which are "cold", those of the tropics can easily be brought under cultivation. In Java and in Sumatra many fens have thus been transformed into rice-, sugar-, and tobacco fields, and can no longer be recognized as fens by the eye. Finally tropical fens are generally much less accessible and a sojourn in such swamps is sometimes rendered quite impossible by large quantities of vermin.

In addition to direct observation, important indicators of fens are the so-called blackwaters, which occur in all parts of the world, where peat-bogs are found. They are known to the Malay people as *ajer itam*, *sungei itam*, *danau itam* etc., but their significance is as little known to geographers as are the fens themselves. The black colour is a result of reflection, as these waters are in reality quite transparent and of the colour of weak tea, the colour being due to the humic acids and alkaline humates derived from the fens.

The following brief survey may afford proof of the wide-spread distribution of fens in the Indian Archipelago, and, in spite of the very incomplete data, we may deduce from it that fens there occupy more than a million hectares.

Java. The largest fen, which is found in Java, is in the valley of Ambarawa (residency Samarang) where its presence was first established by JUNGHUHN. A large portion of it (about 2500 hectares) has been utilized for the cultivation of rice while the remainder (about 1500 hectares) at present still represents the swamp known under the name of Rawah Pĕning. From borings near Fort Willem I it was evident, that this fen has the enormous depth, unknown elsewhere, of 30—31 metres. The Rawah Pĕning further shows itself to be a true fen by the peat islands, which have repeatedly arisen in it, namely in 1838, 1845, 1865, 1868 and 1885.

A fen belonging to the desa Djugelangan, near Kapongan, 15 kilometres east of Panarukan (residency Besuki) has an extent of 21 hectares. The peat made from this contained 27,07% of ash.

Near the desa Tuwiri-wetan, district Djĕnu, division Tuban (residency Rembang) there is a fen with an area of $23^1/_2$ hectares. The peat contains 25,9% of ash.

Peat derived from a fen in the residency Bantam, of which the locality is not further specified, contained 36 % of ash.

South of Meester Cornelis near Batavia several fens are found, of which the Rawah Pondok Tjempaka has been described in detail by H. C. EDELING. In the peat obtained from it 48,5 % of ash was found.

Finally fens are found on the Diëng plateau.

Sumatra. The distribution of fens in the low regions along the north-east coast is very extensive. To these belong in the first place the paja-soils of Langkat, Deli, Serdang etc. (residency Sumatra's East coast) which a comparatively short time ago were still swamps, but have now been reclaimed for tobacco-growing.

The swampy region drained by the Little Siak river which has not yet been examined, probably contains fens, because one of the lakes bears the name Tasik Ajer Itam, and therefore must be counted among the black waters.

The Great Siak river also belong to the black waters. Peat strata have been found along its banks above and below Siak Sri Indrapura. A fen region of about 80.000 hectares lies between the Siak and Kampar rivers. Although known before, it was more closely examined in 1891, at the time of the expedition under J. W. IJZERMAN, by S. H. KOORDERS, especially as regards its flora. There are also extensive fen regions south of the Kampar. The Batang Nila is a black river and one of its affluents is called Ajer Itam.

In Djambi peat strata were found by S. C. CROOKE as early as 1820. In the low countries of Djambi there is an Ajer Itam Laut and in the subdivision Lower Tambesi an Ajer Itam is known.

The residency Palembang is very rich in swamps. In lat. 3°29′ S. and long. 104°55′ E. there is a lake Danau Ajer Itam which is drained by the Ajer Itam. An Ajer Itam Laut has its mouth in lat. 1°20′ S., an Ajer Itam Dalam in lat. 1°13′ S. An Ajer Itam is further found in the subdivision Ogan Ulu and Enim, and another one in the subdivision Ranau districts.

Most fens in Sumatra owe their existence to the fact, that in consequence of the deposit of sludge etc. the beds are constantly raised, so that the banks are also higher than the surrounding country. In the lowest portions of the depressions thus formed between the rivers, lakes and swamps were formed which gave rise to peat formation.

Fens are known in the island *Bengkalis*, and probably it will also be possible to show their presence in the neighbouring islands of *Rupat*, *Padang*, *Rantau* etc. which are also marshy.

In the island of *Bangka* there is an Ajer Itam with a village of the same name, in the district Pangkal Pinang, north of the place of that name.

Borneo. C. A. L. M. SCHWANER was the first to draw attention to the extensive fen formations in the South and East Division. He has further pointed out that the lakes along the Barito and the Kapuwas Murung represent river branches (Danau Kalahin, D. Masura, D.

Madara, D. Ganting, D. Babai, D. Kambat etc.) cut off by constriction, which have become black waters. Similar swamps also occur along the Kahajan. W. J. M. MICHIELSEN found fens along the Sampit and Katingan rivers.

The extensive swamps in the southern part of the Western division in the Kandawangan country have not yet been examined. The names of two rivers, Ajer Itam besar and Ajer Itam ketjil, indicate however that fens are present. This is also the case near Simpang, where the Sungei Itam and the Dulah Sidok belong to the black waters.

The great Kapuwas river is rich in fen formations. Near the mouth a Sungei Itam is found, and swamps occur behind Pontianak. The lakes along this river, which have arisen in the same way as those along the Barito, belong to the black waters. Peat strata were observed along the Kapuwas by G. A. F. MOLENGRAAFF and near the Kenepai by J. E. TEYSSMANN, who also found, that the swamp behind Sintang is a fen. The lake region of the Danau Luwar etc. is also in a state of fen formation. Similarly fens seem to occur along the lower reaches of the Sambas river. A high peat-bog was discovered by G. A. F. MOLENGRAAFF in the Babas Hantu, 1036 metres high, in the Madi mountains.

In Sarawak an extensive swamp was shown to exist by CHARLES HOSE between the Madang and Kalabit districts (lat. 3°16—3°20′ N., long. 115 24—115°38′ E.) and another one in the Western part of the Kalabit district (lat. 3 28′—3°45′ N., long. 115°24′—115°42′ E.). In Northern Borneo there is, according to FRANK HATTON, an extensive fen in the Lamag district, belonging to the basin of the Kinebatangan (lat. 5°13′—5°23′ N., 117°52′—117°56′ E.).

Finally *New Guinea* is very rich in fen formations. Whereas they appear to occur somewhat sparingly in the northern part (Kaptiau on the Borowai and in the territory between the Geelvink Bay and lake Jamur, also near the Sélé Straits), extensive strata occur in the south-western and southern parts of the island. They were found near the Noord-, Reiger-, and Dumas rivers by J. W. VAN NOUHUYS. A sample contained only 4.58% of ash. O. E. HELDRING has traced the distribution in the neighbourhood of Merauke. These investigations extend from the coast to lat. 7°32′ S. and it results from the survey, that of this part 224000 hectares are covered with fens. Many of the rivers which have their mouths on the South coast, such as the Wamal, Bulaka, Bémt, Sewa, Koroi, Bian, Toran, Kumbe, Marau and Torasi should be reckoned among the black waters.

Whereas the composition of the peat-bogs of temperate and cold regions has been subject to many changes, in consequence of the variations

in the climate, there is no trace of this in tropical fens. Accordingly their structure resembles that of the coal measures: just as, in the latter, the roots of Sigillaria and of Lepidodendron — the Stigmarias — are found on the floor of the seams, so the trees of tropical fens take root in the subjacent clay. As these trees continue their growth and are not choked by peat-mosses, and broken as in temperate and cold regions, they frequently remain erect, as is the case with many tree-trunks in the coal measures [1]).

In one respect, however, there is a great difference, because the flora of the carboniferous period has been almost completely modified. The ferns alone, which occur sporadically in recent fens, still remind us vaguely of a remotely distant epoque.

Botany. — "*Some brief remarks relating to the communication of* Professor Dr. C. E. A. WICHMANN *on ""fen formations in the East-Indian Archipelago""*". By Dr. S. H. KOORDERS.

In order to supplement the above communication by Prof. Dr. C. E. A. WICHMANN and also to supplement, what has recently been published on the same subject by Dr. E. MOHR of Buitenzorg and Prof. Dr. J. F. NIERMEIJER of Utrecht [2]), I cannot refrain from briefly communicating here a few short remarks, mainly concerned with the botanical side of the subject.

It appears from a letter from the Königliche Geologische Landesanstalt, dated from Berlin on April 28th last, N°. 5026/A, VIII, that a memoir by Prof. Dr. H. POTONIÉ will soon be published, dealing with the extensive tropical fenformations, discovered in 1891 by the IJZERMAN-expedition in the hot plain of Sumatra; at the time of writing the manuscript was ready for publication.

The principal contents of this letter were as follows:

"Anbei erhalten Sie Ihrem Wunsch gemäss Ihre Bleistiftskizze der mikroskopischen Zusammensetzung des sumatranischen Tropentorfes mit dem verbindlichsten Dank zurück und ebenso den Brief des

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[1]) Since the same tree species which occur in tropical fens, are also found on the surrounding dry land, it is not necessary to regard the Sigillarias, Lepidodendrons and Calamites as bog plants.

[2]) MOHR, Dr. E., in Bulletin du Departement de l'Agriculture Buitenzorg (1908).

NIERMEIJER, Prof. Dr. J. F., Rede bij de aanvaarding v/h Hoogleeraarsambt i/d aardrijkskunde van N. Oost-Indië aan de Rijks-Universiteit te Utrecht (1908).

KOORDERS in POTONIÉ, Prof. Dr. H., Ein von der Holländisch-Indischen Sumatra-Expedition entdecktes Tropenmoor (in Naturwiss. Wochenschrift 20 Oct. 1907 nr. 42, p. 657.

Residenten von Sumatras Ostküste. Ihre mikroskopische Skizze befindet sich in einer Kopie zum klischieren bei unserem Zinkographen zusammen mit den anderen für den Artikel unseres Landesgeologen Professor POTONIÉ bestimmten Figurenvorlagen. Exemplare des Aufsatzes über das sumatranische Tropenmoor im Vergleich mit den Karbonmooren können wir Ihnen noch nicht zustellen, da der im Manuscript vollständig fertig vorliegende Aufsatz erst in Druck gegeben wird, wenn die Klischeeabzüge vorliegen werden. Der Druck soll nach Möglichkeit beschleunigt werden" (msc. 28 April 1909).

The microscopical drawing, referred to in the above letter of the Kgl. Geol. Landesanstalt, was made by me at Leiden on Oct. 31, 1908, as the result of a preliminary microscopical examination, which I undertook immediately after receiving from India the first specimen of peat from the Sumatran "Tropenmoor", referred tc above.

This Sumatran tropical peat was collected at my request on June 29—July 1 1908 by Mr. J. G. LARIVE, civil officer (Controleur) for the division Kamperkiri on the East coast of Sumatra. A copy of the log of Mr. J. G. LARIVE's journey, which I have received, and some data concerning the same district, taken from my own log of March 1891, will be presented by me to the Royal Dutch Geographical Society of Amsterdam, soon after the publication of Prof. POTONIÉ's forth-coming memoir. Meanwhile this memoir will contain a part of a German translation of Mr. J. G. LARIVE's log, which translation was prepared by me and was already sent to Professor POTONIÉ last year.

This tropical peat from the fen formations, discovered in 1891 by the IJZERMAN-expedition in Central Sumatra, was collected at my request and according to my indications by Mr. J. G. LARIVE as the result of an invitation addressed to me by Prof. POTONIÉ in June 1907 at a meeting of the *Botanischen Vereins der Provinz Brandenburg* in Dahlem-Berlin.

My request was transmitted to Mr. LARIVE through the much-valued agency of General G. M. BLECKMANN (at that time colonel of infantry at Soerabaja).

As I naturally wish to postpone my detailed communication to the Royal Dutch Geographical Society until Prof. POTONIÉ will have published his investigation of the tropical peat specimens, I limit myself here to the above brief remarks.

Meanwhile I wish to express my hearty thanks to Mr. J. G. LARIVE, who quite disinterestedly and on behalf of science was good enough to undertake the labour of a very difficult journey. In accordance with my wish, Mr. LARIVE collected the specimens personally at the

same spot, where I found a fen stratum of 8 metres thickness, on March 20—21 1891 at the time of the IJzerman-expedition.

At the same time I tender my hearty thanks to my friend, General G. M. Bleckmann, for his kind assistance, by which my request was transmitted to Mr. J. G. Larive through the valuable intervention to the Resident of Sumatra's East coast.

Leiden, May 29th 1909.

Physics. — "*On Phosphorescence at very low temperatures.*" By Henri and Jean Becquerel, and H. Kamerlingh Onnes. Communication N°. 110*a* from the Physical Laboratory, Leiden.

(Communicated in the meeting of April 23, 1909).

§ 1. *Introduction.* It is known that in the case of the salts of the rare earths both in their crystalline form and in solution, absorption spectra with particularly clear and fine lines can be obtained by the application of very low temperatures, and that the investigation of the optical and magneto-optical properties of these substances at the temperatures which can be reached by means of liquid air and liquid hydrogen opens up a new method for the study of selective absorption [1]).

A problem, not less important than this, is that of the emission of light at temperatures below that of beginning incandescence. For certain substances, such as the *uranylsalts* [2]), there exists such an intimate connection between the bands in the emission spectra of their phosphorescence and the bands of their absorption spectra, that their distribution in each spectrum follows the same law, so that the bands in the one spectrum form the continuation of those in the other, while they have even two bands in common. Any peculiarity in the occurrence of one of the bands in any of the compounds periodically recurs in all the other bands, as well in the emission as in the absorption spectrum.

Thus, when the absorption spectrum of any of the uranyl compounds

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1) Jean Becquerel Compt. Rend. 1906, 1907, 1908, 1909. le Radium t. IV. p. 49, p. 107, p. 328, p. 383. t. V p. 5; Physik. Zeitschr. t. VIII p. 632, p. 929. Jean Becquerel and H. Kamerlingh Onnes, These Proc. Febr. 1908. p. 592. Comm. fr. the physic. Labor. at. Leiden n°. 103. Compt. Rend. t. CXLVI p. 625. le Radium t. V p. 227.

2) Edmond Becquerel Mém. de l'Ac. d. Sciences t. XL. Ann de Ch. et de Phys. 3e ser t. X p. 5. Henri Becquerel Compt. Rend. t. CI 1885 p. 1252.

changes when brought under the influence of a very low temperature, the same change is also to be expected in the emission spectrum of its phosphorescence. This expectation is fully supported by our experiments. The more or less broad and diffuse bands which follow each other in regular sequence in the spectra of the uranyl salts at ordinary temperature, are, at the temperature of liquid air, resolved into multiple bands, which, in some cases, are particularly fine and intense; in that respect, all the bands in both emission and absorption spectra undergo identical changes. The appearance of these spectra at liquid air temperature has already been described [1]). At the Leiden cryogenic laboratory we have extended the investigation of these phenomena to the temperature of solid hydrogen (14° K.), and the most important of the results thus obtained concerning emission spectra at very low temperatures are here published.

§ 2. *Experimental method.* The apparatus here used for the investigation of the spectra of phosphorescence was the same as that which had already served for the investigation of absorption spectra, and for a description of which we refer to former papers [2]).

Here we have only to mention that the source of the spectrum was a ROWLAND plane grating with a lens of 1.30 m. focal length arranged in auto-collimation, and that the phosphorescent substances, enclosed in thin-walled glass tubes, were placed in a non-silvered vacuum vessel containing liquid hydrogen; in the latter case the glass containing liquid hydrogen was placed in a second vacuum vessel with liquid air so as to reduce the vaporisation of the hydrogen. The light of an arc-lamp, filtered through an ammoniacal solution of copper sulphate, served to illuminate the salts.

§ 3. *Behaviour of the spectra when the temperature is lowered. The displacement and the limiting position of the phosphorescence bands of uranyl salts.*

When uranyl salts are cooled to a temperature of 14° K. (solid hydrogen), the lustre which they show at ordinary temperature is not diminished and vanishes with the illumination. Hence, *lowering of the temperature of uranyl salts in no way prevents nor prolongs or shortens* [3]) *their emission of light.*

The bands which, between ordinary temperature and that of liquid air already undergo subdivision, become groups of much finer lines at the temperature of liquid hydrogen.

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[1]) HENRI BECQUEREL Compt. Rend. t. CXLIV 1907 p. 459 and p. 671.

[2]) Proc. 29 Febr. 1908; Comm. Phys. Lab. Leiden N°. 103. Le Radium t. V, p. 227.

[3]) Added in the translation

Fig. 1 which gives a part of the spectrum of the double sulphate of potassium and uranyl at temperatures of 288° K (ordinary temperature), 80° K. (liquid nitrogen) and 20° K. (liquid hydrogen), shows this very clearly. The group in the bluish green is very weak, but three groups of bands in the green are very strongly developed. The manner in which the broad, diffuse patches at ordinary temperature are resolved at the lower temperatures into groups of bands is here strikingly shown. Figs. 4 and 5 (obtained with the second spectrum of the grating) give the details of some groups of bands of the same salt. Some bands which still remained complex at 80° K. have been resolved between 80° and 20°K. The spectra *a* and *b* of fig. 1 were obtained alongside each other upon the same plate by projecting the image of the phosphorescent salt upon two different parts of the slit in succession. From them we can see that *by lowering the temperature the emission-maxima are displaced towards the smaller wave-lengths.* This has already been observed [1]), but it was not then shown that the displacement of the maxima was the result of a change in the period of vibration; it could also have been the result of an unusual strengthening of bands which existed already at ordinary temperature but had then a very small intensity. The observation of the spectrum at temperatures at which the bands are distinctly separated from each other shows that we have here to deal with a displacement resulting from the lowering of the temperature. The following measurements, obtained by comparison with the iron spectrum, show a distinct displacement of the bands in the case of the double sulphate of uranyl and potassium between 80° K and 20° K.

80° K.	511.48 $\mu\mu$	534.24 $\mu\mu$	559.09 $\mu\mu$	586.31 $\mu\mu$
20° K.	511.35	534.10	558.89	586.05.

Between 80° K. and 20° K. the ratio of the displacement to the temperature difference expressed in degrees is much smaller than between ordinary temperature (288° K.) and 80° K. (It is then from 2 to 3 $\mu\mu$).

The displacements obtained by further lowering of the temperature from 20° K. to 14° K. are scarcely noticeable (fig. 5 *a* and *b*), as was to be expected by extrapolation paying attention to the small difference in degrees between these temperatures. The order of magnitude is from 0,01 to 0,02 $\mu\mu$. Hence, *it is very possible that as the temperature is lowered, the bands approach asymptotically a limiting position.* Their change, therefore, is of the same nature as the change of volume and, perhaps, as the change of dielectric constant.

[1]) Henri Becquerel, Compt. Rend. t. CXLIV 1907 p. 671.

§ 4. *Comparison of the different groups of bands of the same salt.*

The photographs of Pl. I and II show better than any description the spectra of a number of uranyl compounds. The data for the separate spectra are given with the explanation of the figures.

The spectrum of a definite salt consists of different groups (we observed from 7 to 8), which though not quite the same, still show a very small difference between successive groups. Only the most refrangible group, some bands of which, as we shall see later, belong both to the emission and absorption spectrum, is somewhat different from the others. We shall call those bands homologous which occupy the same relative position in the different groups. Such, for instance, are the bands whose wave-lengths are marked on fig. 7. (Double acetate of uranyl and sodium).

The difference in the appearance of the different groups is the result of successive ascending changes in the relative intensities of the bands in these groups. This can be seen in all the figures, especially in fig. 7. The band 566$^{\mu\mu}$.34 (fig. 7) is the most intense in the least refracted group visible in the figure; in the groups succeeding this one towards the side of the smaller wave-lengths, the homologous bands 540.$^{\mu\mu}$45, 516.$^{\mu\mu}$78, 495.$^{\mu\mu}$02 become steadily weaker, and in the blue group the band which should be homologous with the foregoing seems to have disappeared.

On the other hand, the bands which occur on either side of the above-mentioned homologous series, preponderate in the more refracted portion of the spectrum, but become less distinct as the greater wave-lengths are approached.

The spectra of uranyl sulphate has the same general appearance quite independently of whether it is combined with other sulphates or not. (See figs. 1, 8, 9, 10). It was noticed long ago [1]) that the structure of the spectrum depends chiefly upon the acid with which the salt is formed, and is little influenced by the other base in the double salt. But this is shown much more clearly by observations at temperatures at which the bands no longer overlap.

§ 5. *Law of succession of the bands.* One of the most important problems is to ascertain the law connecting the frequencies of successive bands. Observations at ordinary temperature had already shown that the broad bands (such as those of fig. 1*a*) were at such distances apart, that the difference between the frequencies of successive

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[1]) Edmond Becquerel, Mém. de l' Acad. d. Sc. t XL p. 8 and 15.

TABLE.

Double sulphate of uranyl and potassium.

Series in which the strongest bands occur, at 20° K.

λ	490.34 μμ	— 511.35 μμ	— 534.10 μμ	— 558.89 μμ	— 586.05 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	837	833	830	829	

The same series at 80° K.

λ	490.42 μμ	— 511.48 μμ	— 534.24 μμ	— 559.09 μμ	— 586.31 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	836	833	831	830	

Double sulphate of uranyl and sodium.

Series of strong bands 80° K.

λ	489.15 μμ	— 510.12 μμ	— 533.00 μμ	— 557.85 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	840	841	835	

Another series of strong bands at 80° K.

λ	512.54 μμ	— 535.47 μμ	— 560.46 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	837	833	

Double sulphate of uranyl and ammonium.

Series of strong bands at 80° K.

λ	491.26 μμ	— 512.45 μμ	— 535.54 μμ	— 560.74 μμ	— 588.11 μμ	— 618.40 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	842	841	839	829	833	

Another series of strong bands at 80° K.

λ	493.47 μμ	— 514.78 μμ	— 538.01 μμ	— 563.33 μμ	— 590.80 μμ	— 621.57 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	839	839	835	825	838	

Uranylsulphate.

Series of strong bands at 80° K:

λ	491.83 μμ	— 513.39 μμ	— 536.94 μμ	— 562.68 μμ	— 591.01 μμ	— 621.95 μμ
$\Delta \frac{1}{\lambda} \times 10^7$	854	853	852	852	844	

Another series of strong bands at 80° K.

λ	516.02 $\mu\mu$	539.58 $\mu\mu$	565.40 $\mu\mu$	593.88 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$		843	850	848

Uranylnitrate.

Series of the strongest bands at 80° K.

λ	506.96 $\mu\mu$	530.15 $\mu\mu$	555.46 $\mu\mu$	583.29 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$		862	859	858

Double acetate of uranyl and sodium.

Six series at 80° K.

λ	473.25 $\mu\mu$	493.25 $\mu\mu$	514.86 $\mu\mu$	538.45 $\mu\mu$ — 564.27 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$	855	852	851	850

λ	495.02 $\mu\mu$	516.78 $\mu\mu$	540.45 $\mu\mu$	566.34 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$		851	848	846

λ	476.23 $\mu\mu$	496.49 $\mu\mu$	518.49 $\mu\mu$	542.40 $\mu\mu$ — 568.58 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$	857	855	849	849

λ	476.92 $\mu\mu$	497.21 $\mu\mu$	519.22 $\mu\mu$	543.16 $\mu\mu$ — 569.46 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$	856	853	848	850

λ	478.58 $\mu\mu$	499.02 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$	856	

λ	479.46 $\mu\mu$	499.92 $\mu\mu$	522.14 $\mu\mu$ — 546.35 $\mu\mu$
$\Delta \frac{1}{\lambda} \times 10^7$	854	851	849

bands was almost constant[1]). But as the bands at this temperature are broad, diffuse, and complex, it was not possible to determine the accuracy of the relation. In the spectra obtained at low temperatures the wave-lengths could be measured; this was done by comparison with the iron-spectrum which was photographed upon the same plate as the phosporescence spectrum (see, for instance, figs. 6, 7, 8, 10).

A high degree of accuracy is possible in these measurements, particularly at the temperature of liquid hydrogen: in the case of the double sulphate of uranyl and potassium (fig. 6) and of the double acetate of sodium and uranyl (fig. 7) the error in the position of the principal bands is not more than $\pm 0^{\mu\mu},03$. The table given on pp. 80 and 81 contains the results of the measurements for the bands of certain salts, homologous bands being placed upon the same line.

The values of wave-lengths given above were measured in air, but the differences between the reciprocal wave-lengths are reduced to vacuum, so that the values given for these differences are proportional to the frequencies.

From the table it appears that *the difference between the frequencies of two successive homologous bands is practically constant, not only for the same, but also for all series of homologous bands of the same salt. Further, the values of this constant for the various salts differ but slightly from each other.*

As regards the position of the bands, the spectrum of a uranyl salt is thus represented on the scale of frequencies by successive identical groups at an equal distance from each other. The deviations from this law are, as a general rule, so small, that they can be ascribed to errors in the measurements of the wave-lengths; nevertheless there is a tendency towards decrease in the frequency-differences with increasing wave-length, and it is possible, and even probable, that there is a deviation from the law in this direction, although in that case, the deviation is extremely small.

In the foregoing we have recalled the influence of the acid upon the structure of the spectrum; now we must call attention to the fact that although these spectra are also *spectra of molecules*, which differ in the various salts, yet their character is due to the uranium, which subjects the spectra of all the salts to the law just mentioned.

The law according to which the homologous bands correspond with equal differences between the frequencies is the simplest that could connect successive lines in the spectrum.

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[1]) Edmond Becquerel loc. cit. § 4 etc. Henri Becquerel Compt. Rend. t. CI 1885 p. 1259.

In this connection we may recall that this law, regarded as holding with close approximation, had long ago drawn attention to the uranyl salts in particular as being, from the point of view of optical phenomena, of remarkable molecular structure. This formed the starting point of the first experiments which were undertaken with a view to discovering rays analogous to those which had been observed in vacuum tubes. [1]) They led to the discovery of radioactivity.

§ 6. *Character and nature of the phosphorescence spectra of the uranyl salts.*

We have seen what the law is that governs the succession of the homologous bands in the various groups. Is, now, the disposition of the bands in the same group governed by a particular law? In the most intense portion of the green of the spectrum the bands are numerous, especially at the temperature of liquid hydrogen, forming a system in themselves. But regarding the bands at the ends of the spectrum, we find, especially in the last group in the red, that they become simpler, and in many cases there remains only a succession of bands lying close together and showing marked similarity with the groups of bands in the channeled spectra of gases (nitrogen, carbon) when these are observed with a weak dispersion. In the least refrangible groups in the spectrum of uranyl sulphate (fig. 10) this is particularly well marked. This group consists of a strongly developed head towards the side of the small wave-lengths followed by seven or eight bands at regular distances from each other, and decreasing regularly in intensity as they go further way from the head. Each of these bands is a little asymmetrical, for the side towards the smaller wave-lengths is more sharply defined than the other. The blue group in the spectrum of the same salt (fig. 11) is also a channeled spectrum, but it is not so extended and seems to be formed out of various series of overlapping bands.

The bands are not sufficiently fine and especially they are not sufficiently numerous for us to examine if the successive bands in the same group and the initial bands in the various groups follow DESLANDRES' law. Nevertheless this law receives as much support as might be expected especially from the groups in the orange and red in the case of the simple sulphate.

It seems to us, therefore, that these spectra are channeled spectra of the same character as the band spectra of gases.

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[1]) HENRI BECQUEREL, Compt. Rend. CXXII, p. 420. (1896).

We have investigated if a magnetic field has any influence upon the spectra of the uranyl salts. *In a field that could exceed* 20 *Kilogauss in value, no influence even upon the narrow bands at* 14° *K.* (*solid hydrogen*) *was to be observed.* This negative result forms a new analogy between these spectra and the band spectra of gases, which likewise, in the majority of cases undergo no change in a magnetic field, although the strength of this is raised to the highest value at present possible [1]).

§ 7. *Relation between the emission by phosphorescence and the absorption of the uranyl salts. Reversible and irreversible bands.*

In the foregoing we have recalled the existence of groups which contain at the same time emission and absorption bands. Crystals of *autunite* (double phosphate of uranyl and calcium), a mineral from which very transparent plates may be obtained by cleavage, are especially suitable for the study of absorption.

Fig. 12 shows the emission and absorption spectra in the blue and green region of the spectrum of a plate of autunite at a temperature of 14° K. (solid hydrogen). It can be seen that two bands lying very close beside each other are common to both spectra; α is well developed both as an emission and absorption band, β is strong as an emission and weak as an absorption band.

Fig. 13 gives the spectrum of autunite at 80° K.

In can be shown experimentally in a striking manner that some bands can arbitrarily become emission or absorption bands. For this purpose an intense beam of light is transmitted through the crystal plate and the position of the absorption band fixed; the intensity of the transmitted light is then diminished and at the same time the plate is illuminated by violet light whose intensity always increases. *The dark band is seen to be transformed into a light band occupying exactly the same position.* This experiment, which becomes possible at the lowest temperatures, resembles the classical experiment of the reversal of the lines in the case of the sodium-flame.

The bands in the less refrangible groups cannot be reversed; they all belong to the emission spectrum.

The marked difference between the groups which are common to both spectra, and those which belong only to the emission spectrum, can be attributed to the reversible bands whose relative intensity,

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[1]) This, however, is not generally the case. As is known, A. DUFOUR has found numerous bands in the spectra of the chlorides and fluorides of the alkaline earths which show a ZEEMAN effect, and one of us has also observed this effect in the yttrium-bands.

compared with that of the other emission bands, is much modified by this property.

The experiment concerning the reversal of certain bands of the uranyl salts is the first proof that certain emission bands occupy *exactly* the same position as the absorption bands. Further, the irreversible homologous bands of the other groups correspond with vibration-frequencies of the absorbed light separated from these bands by constant differences.

There seems, therefore, to exist an immediate and simple relation between absorption and emission in the uranyl salts.

We may recall that the uranosalts, *which do not phosphoresce*, have absorption spectra in which the bands with marked regularity follow the law of succession of the emission bands of the phosphorescent uranyl compounds [1]).

§ 8. *Phosphorescence of the sulphur compounds at low temperatures.*

We have made but very few observations of the phosphorescence at low temperatures of phosphorescent substances other than the uranyl salts. As regards sulphur compounds of the alcaline earths we could add nothing to the masterly research of LENARD and KLATT [2]).

Zinc sulphide is very luminous when excited by violet light at 80 K. and remains luminous for a long time afterwards; 4 or 5 seconds after excitation ceases, the stronger radiation comes to an end, and then for a long time afterwards the zinc sulphide shines with a soft glow. From the moment that warming begins as it returns to ordinary temperature, zinc sulphide shows a particularly strong lustre.

At hydrogen temperatures the light emitted during excitation, and by warming after excitation, becomes less.

The colour of the light coming from zinc sulphide seems to be the same at various temperatures, and between 80° K. and ordinary temperature no change can be seen in the spectrum, which was not unexpected seeing that the spectrum was continuous.

§ 9. *Conclusion.* The uranyl salts must be regarded as a class by themselves sharply differentiated from other classes of phosphorescent substances.

From the researches of LECOCQ DE BOISBAUDRAN, LENARD and KLATT, and URBAIN on the rare earths, the sulphur compounds of the alkaline

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1) HENRI BECQUEREL Compt. Rend. t. CI. 1885, p. 1252.

2) P. LENARD and V. KLATT, Ann. d. Phys. IV. 15. 1904, p. 225—282 and 425—672.

earths, and the oxides of the earth metals, it is known that these substances in the pure state do not phosphoresce; and it appears that the influence of impurity, as was first noticed by EDMOND BECQUEREL, is essential to the formation in those substances of complex molecules, LENARD's "centra", which, as distinguished from the molecules of the substances themselves, have the power of emitting light.

It is completely different with the salts of uranyl. These compounds always phosphoresce, even when quite pure, and each of them is characterised by a specifically distinct emission spectrum.

And in other respects the uranyl salts differ from those just mentioned as phosphorescent only by the influence of admixture. Even at a temperature so low as that of liquid hydrogen, in intensity the emitted light does not alter and vanishes with the illumination [1]). The uranyl salts do not exhibit thermoluminescence. Finally, the spectra of these salts are of remarkable structure, while an immediate relation exists between the emission and absorption spectra.

These optical phenomena at low temperatures, by which considerable light is thrown on the characteristic properties of the phosphorescence of uranyl salts, thus lead to a distinction between two kinds of phosphorescence, which seem to be of different origins. On the one hand, in the case of the rare earths, the sulphur compounds of the alkaline earths, and the oxides of the earth metals, according to the beautiful theory of LENARD [2]), the electrons, which, under the photo-electric influence of the absorbed light, are ejected by the atoms of the metal and by their return to these cause phosphorescence are supplied by that metal of which a small quantity is added to the experimental substance so as to cause it to phosphoresce by the formation of "centra", of which the structure of the molecular complexes play an important part in the phosphorescence. On the other hand, following in the track of the same theory, the explanation of the phenomena exhibited by uranyl salts must be sought in the motion of the electrons contained in the uranium atom itself and in the structure of the molecules of the substance itself.

In this case especially considering the small obstruction cold offers to the return of the electrons [3]) the question obtrudes itself if, perhaps, the whole process of phosphorescence takes place within the uranium atom, or at least within the uranyl group, and electrons, under the photo-electric influence of the absorbed light, are projected from a part of this atom to other parts, remain there temporarily, and, by

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[1]) Added in the translation.

[2]) P. LENARD. Verh. d. Naturhist. Med. Ver Heidelberg 5 Febr. 1909.

[3]) The emission vanishing with the illumination. (Added in the translation).

Fig. 1.

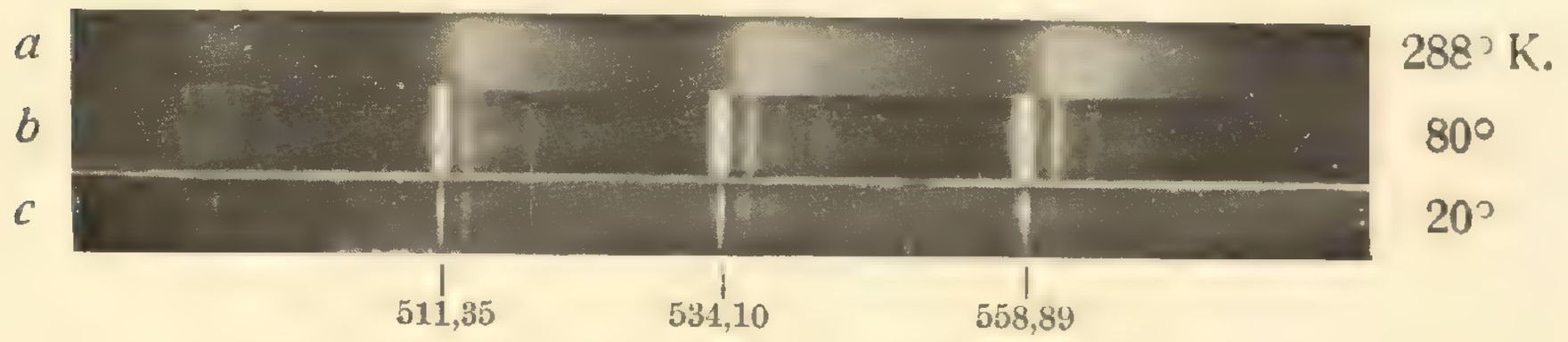

Fig. 2.

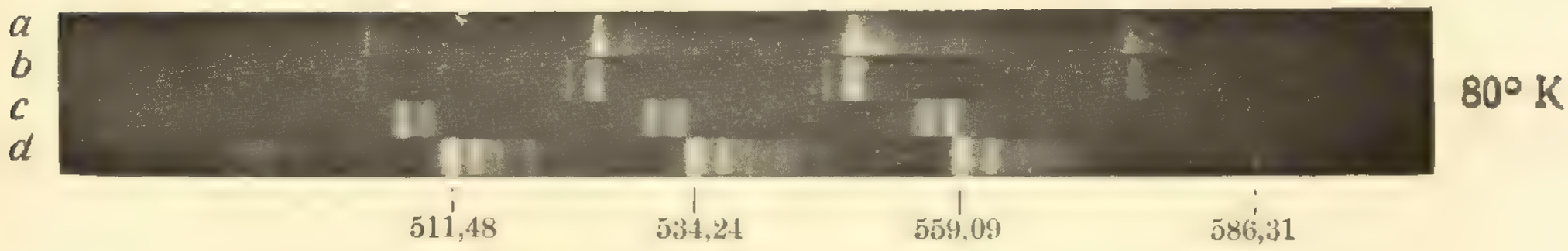

Fig. 3.

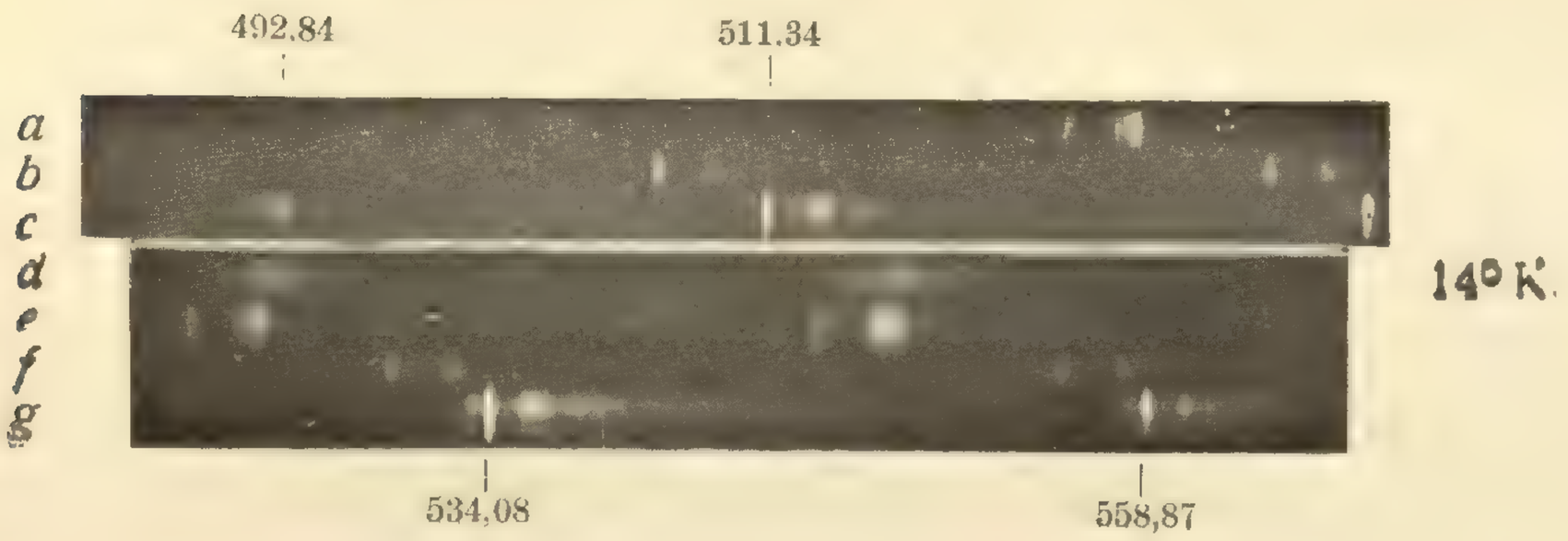

Fig. 4.

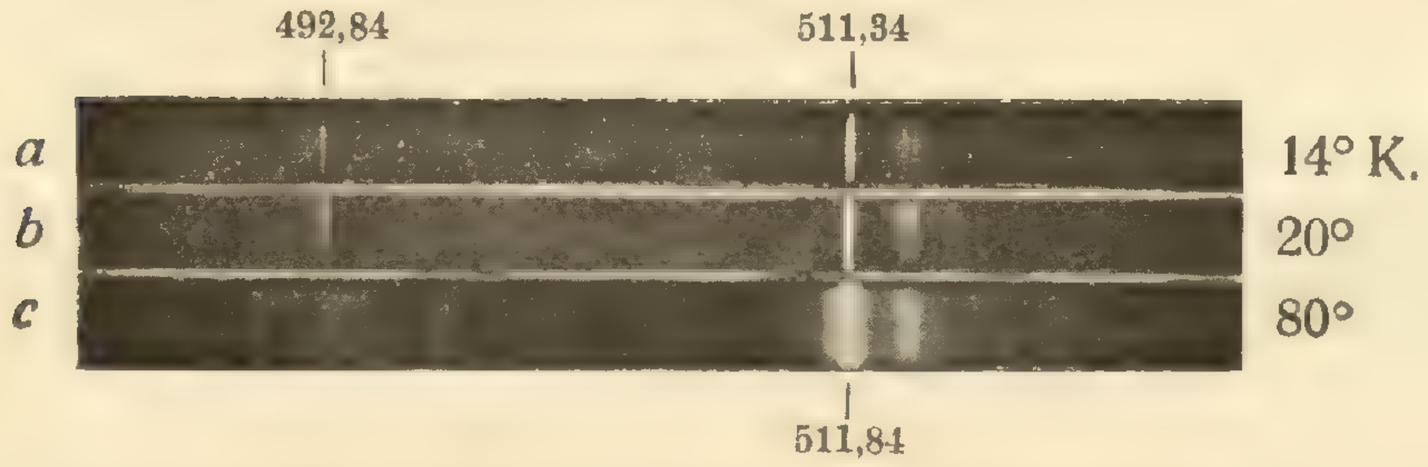

Fig. 5.

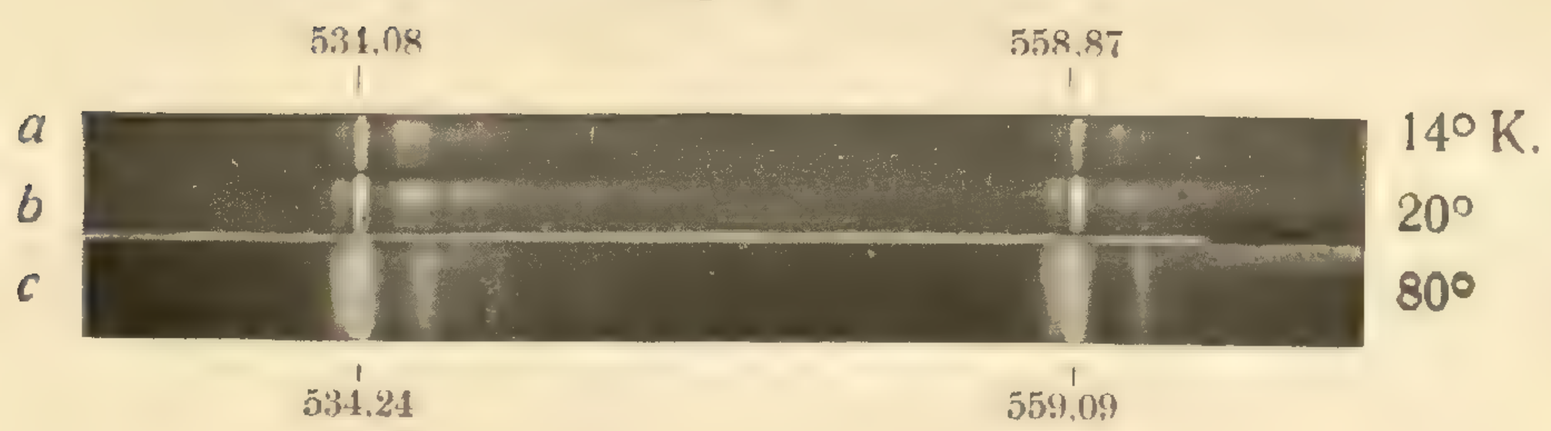

Fig. 6.

Fig. 7.

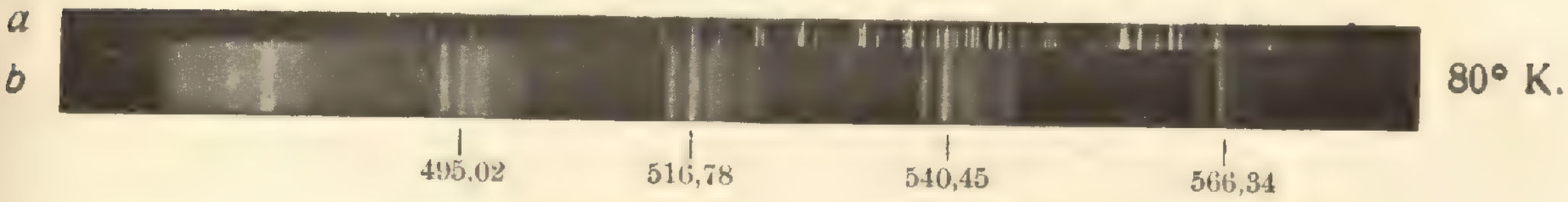

Fig. 8.

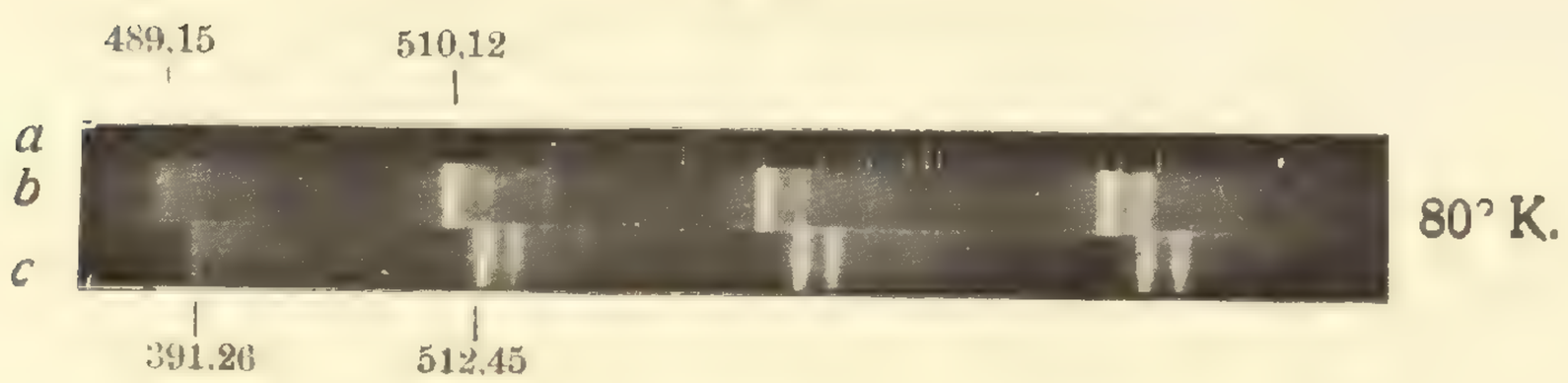

Fig. 9.

Fig. 10.

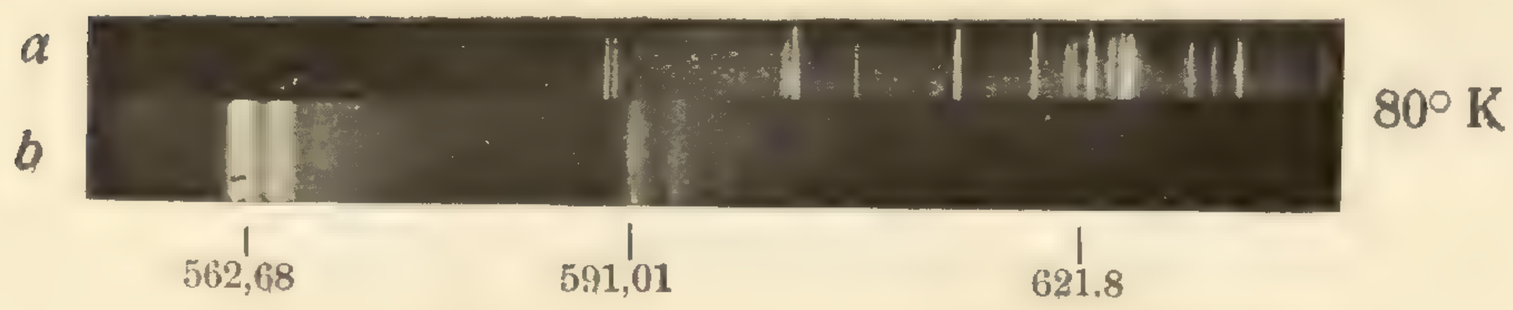

Fig. 11.

Fig. 12.

Fig. 13.

returning to their original position of equilibrium, cause the emission of phosphorescent light. The motion of the electrons causing phosphorescence, and therefore, the position of the bands in the spectrum will, of course, undergo different modifications for the various salts by the influence (of the electric field) of the other atoms, which with the atom of uranium form a complete molecule.

EXPLANATION OF THE FIGURES.

Fig. 1. *Potassium uranyl sulphate* *a*) at 288° K, *b*) at 80° K, *c*) at 20° K. First spectrum, ROWLAND grating.
The spectra *a*) and *b*) photographed beside each other on the same plate allow a direct comparison of the positions of the bands at ordinary temperature and at that of liquid nitrogen.

Fig. 2. *a*) *Autunite* (Calcium uranyl phosphate).
b) *Potassium uranyl chloride.*
c) *Uranyl nitrate.*
d) *Potassium uranyl sulphate.*
Temperature 80° K. First spectrum, ROWLAND. grating.

Fig. 3. *a*) and *c*) *Potassium uranyl chloride.*
b) and *f*) *Uranyl nitrate.*
g) *Potassium uranyl sulphate.*
d) *Autunite* (calcium uranyl phosphate).
Temperature 14° K. Second spectrum, ROWLAND grating. These photos give the details of the groups at the temperature of solid hydrogen.

Fig. 4 and 5. *Potassium uranyl sulphate.*
a) at 14° K., *b*) at 20°K., *c*) at 80°, K.
Second spectrum, ROWLAND grating. Details of the groups at these three temperatures.

Fig. 6. Adjacent spectra of *a*) *potassium uranyl sulphate* at 20° K. and *b*) *the iron arc.*
Second spectrum, ROWLAND grating. Example of the measurement of wave-length; short exposure so as to make an accurate measurement possible.

Fig. 7. *Sodium uranyl acetate* at 80° K.
First spectrum, ROWLAND grating.
Measurement of wave-lengths, and details of the groups.

Fig. 8. *a*) *Iron arc, b*) *Sodium uranyl sulphate, c*) *Ammonium uranyl sulphate.*
Temperature 80° K.
First spectrum, ROWLAND grating.
Measurement of wave lengths, and details of the groups.

Fig. 9. *Uranyl sulphate* at 80° K.

Fig. 10. *a) Iron arc, b) Uranyl sulphate.*
Temperature 80° K.
First spectrum, ROWLAND grating.
Channeled spectrum in the yellow and orange.

Fig. 11. *Uranyl sulphate.*
Temperature 80° K.
Second spectrum, ROWLAND grating.
Channeled spectrum in the green-blue.

Fig. 12. *Absorption and emission of autunite* (calcium uranyl phosphate) at 14° K.
Second spectrum, ROWLAND grating.
a) emission spectrum, *b*) absorption spectrum, α and β, bands common to both spectra (reversible bands).

Fig. 13. *Absorption and emission of autunite* at 80° K.
First spectrum, ROWLAND grating.
a) and *b*) two of the chief absorption spectra of the crystal plate, *c*) phosphorescence spectrum, α) reversible band.

ERRATUM.

In the Proceedings of the Meeting of Oct. 31, 1908.

p. 335 line 5 from the bottom: for Pt'_d read Pt_d.

the head of Table I read: Comparison between Pt'_l and Pt''_d.

p. 341 line 1 from the bottom: for 1807 read 1907.

(June 25, 1909).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Saturday June 26, 1909.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van Zaterdag 26 Juni 1909, Dl. XVIII).

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Physiology. — *"About Odour-affinity, based on experiments of* J. HERMANIDES. By Prof. H. ZWAARDEMAKER.

(Communicated in the meeting of May 29, 1909).

In the physiological laboratory at Utrecht Mr. HERMANIDES prepares a dissertation about the constants of olfactometers used in qualitative measurement of odour [1]). The latter are nine in number. The odorous matter consists as much as possible of pure substances, taken up by paraffinum liquidum, with the exception of muscon, which is divided in myristine-acid. In the measurements first of all the weakest observable stimulus is fixed for each olfactometer separately. In order to compare the results that Mr. HERMANIDES arrived at, with those of other observers we give in the subjoined table a review of minimum values of stimuli. Each figure represents the mean out of 10 observations.

MINIMA PERCEPTIBILIA

in cm.

	Z_1	Z_2	N	K	H_5	v. d. H. L. abnormal odour-syst. [2])	Ha
Isoamylacetate $^1/_2$%	0.23	0.2	0.3	0.245	0.3	0.34	0.4
Nitrobenzol 5 % .	0.03	0.03	0.03	0.02	0.02	0.007	0.03
Terpineol 2.5% . .	1.10	1.10	2.0	2.90	1.3	>7.00	1.2
Muscon 0.627% . .	0.15	0.15	0.3		0.2	1.45	0.3
Aethylbisulfide 1‰	0.015	0.015	0.005	0.004	0.012	0.28	0.02
Guajacol 1‰ . . .	0.30	0.30	0.25	0.50	0.5	2.30	0.6
Valerian acid 1‰	0.03	0.03	0.02	0.02	0.04	0.05	0.04
Pyridine 1‰. . . .	0.02	0.02	0.02	0.023	0.03	2.25	0.03
Skatol 1‰.	0.002	0.003		0.0003	0.002	0.000043	0.003

The names of the observers whose initials are given at the head of the columns, are: ZWAARDEMAKER, NOYONS, KUBO, HERMANIDES, VAN DER HOEVEN LEONHARD, HERINGA. The mean error in the observations of Mr. HERMANIDES was very satisfactory, viz.:

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[1]) See Meeting 28 Sept. 1907.

[2]) At the same time abnormal trichromate, as to sense of colour; compare Onderz. Physiol. Lab. Utrecht (5), Deel. 8, p. 394.

for	the	isoamylacetate-olfactometer		16 %
,,	,,	nitrobenzol-	,,	40 ,,
,,	,,	terpineol-	,,	20 ,,
,,	,,	muscon-	,,	24 ,,
,,	,,	aethylbisulfide-	,,	10 ,,
,,	,,	guajacol-	,,	20 ,,
,,	,,	valerianacid-	,,	18 ,,
,,	,,	pyridine-	,,	12 ,,
,,	,,	scatol-	,,	23 ,,

Expressed in absolute measure the minimum perceptibile of HERMANIDES answers to:

for	isoamylacetate	9.10^{-5}	grammes per Liter of air
,,	nitrobenzol	412.10^{-7}	,, ,, ,, ,, ,,
,,	terpineol	185.10^{-6}	,, ,, ,, ,, ,,
,,	aethylbisulfide	3.10^{-7}	,, ,, ,, ,, ,,
,,	guajacol	37.10^{-7}	,, ,, ,, ,, ,,
,,	valerian acid	21.10^{-7}	,, ,, ,, ,, ,,
,,	pyridine	1625.10^{-7}	,, ,, ,, ,, ,,
,,	scatol	35.10^{-8}	,, ,, ,, ,, ,,

Mr. HERMANIDES has joined together his 9 standard odours in all possible combinations and tried a compensation. By alternately rendering one of the two stimuli weaker or stronger he determined the quantitative proportion, for which the blended odour either entirely disappeared, or approached to zero in the double olfactometer, first in case of unilateral, afterwards also with bilateral supply. That the blended odour approached to zero, was assumed to be the case, when in the combination only an extremely weak indefinite odour was observed. The whole number of combinations each of 2 odours examined in this way amounted to 72. Each mixture repeating itself twice in unilateral observation, 36 combinations were thus formed. The proportional numbers met with for these, are taken together in the subjoined table, whilst the table taken from my first communication regarding odour-affinity, to render a comparison possible, has been placed by its side. The odour-stimuli are expressed in olfactions and each time the proportion $\frac{p}{q}$ is given, in which for each combination the quantities had been mixed.

PROPORTIONAL NUMBERS OF THE STIMULI COMPENSATING EACH OTHER.

ZWAARDEMAKER.

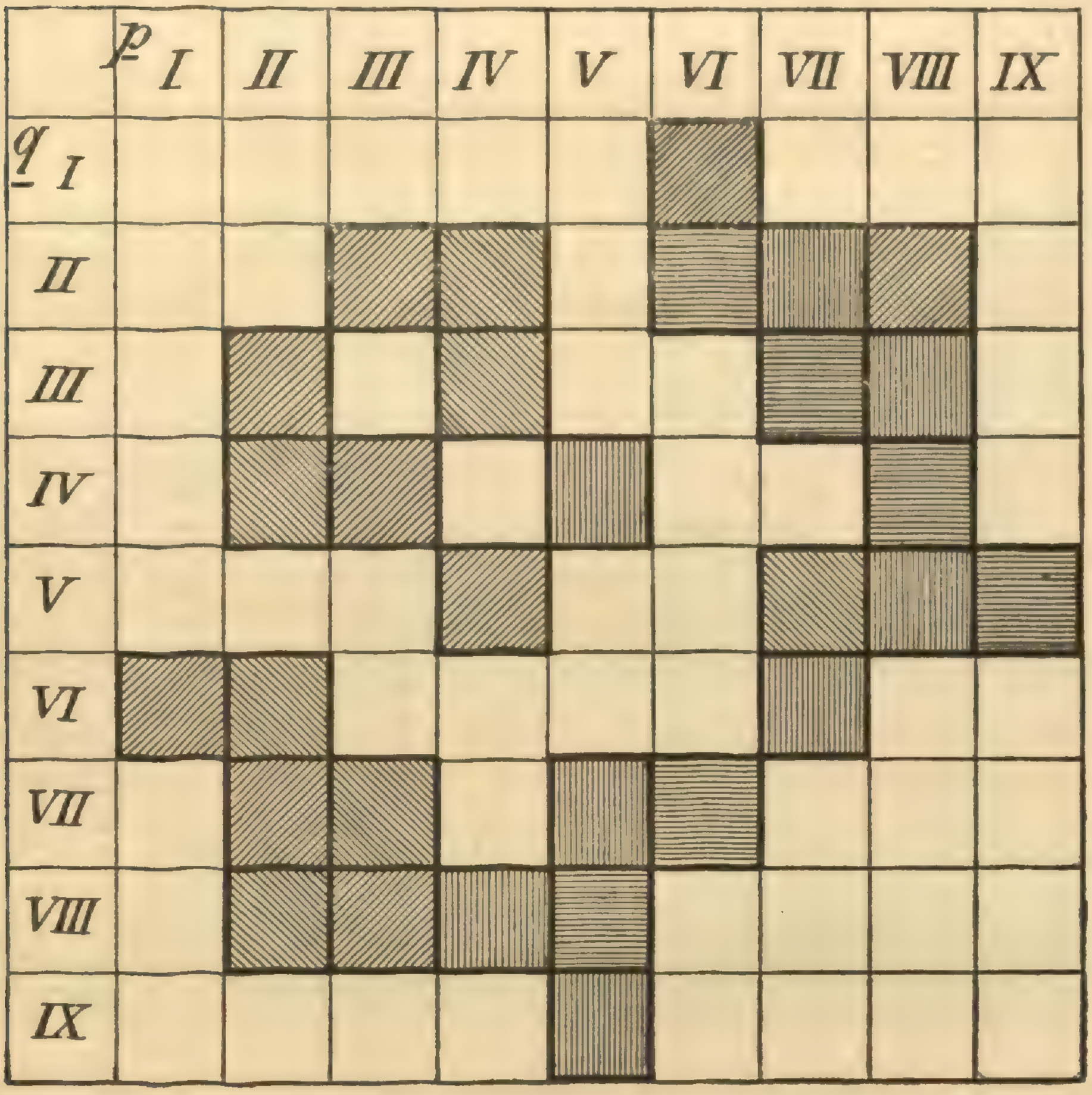
q \ p	I	II	III	IV	V	VI	VII	VIII	IX
I		2.2	0.76	16	4.1	0.33	107	0.28	375
II	0.44		0.73	2.3	9	1.53	30	0.33	83
III	1.32	1.37		8	14	1	20	2	8
IV	0.06	0 43	0.12		1	0 03	0.33	0.83	5
V	0.24	0.11	0.07	1		0.06	0.82	0.31	44
VI	3	0.65	1	33	18		33	63	1400
VII	0.01	0.03	0.05	3	1.2	0.03		0.25	0.08
VIII	3.6	3	0.5	1.2	3.2	0.02	4		2.4
IX	0.003	0.012	0.125	0.2	0.023	0.001	12	0.42	

HERMANIDES.

q \ p	I	II	III	IV	V	VI	VII	VIII	IX
I		1.18	0.26	5.26	1.25	0.5	15.07	0.16	18
II	0.85		0.45	1.24	0.54	1.64	70.4	0.25	5.2
III	3.88	2.22		5.5	2.22	2.94	16	1.74	22.86
IV	0.19	0.81	0.18		0.44	0.37	1.61	1.82	24
V	0.8	1.87	0.45	2.27		0.52	1.72	0.54	27.03
VI	2	0.61	0.34	2.69	1.92		28.57	0.67	18.52
VII	0.063	0.0126	0.062	0.62	0.58	0.035		0.036	3.2
VIII	6.15	3.99	0.58	0.55	1.85	1.5	27.39		23.26
IX	0.055	0.19	0.04	0.042	0.037	0.054	0.31	0.043	

In order to give a survey of the proportional numbers in which the two observers agree and in which they differ, the cases in which there is agreement within the limits of the probable errors, are expressed in the subjoined field of squares.

Cases in which the proportional number $\frac{p}{q}$ does not differ in Z and Hs, within the limits of the probable errors.



A definite order in these agreements is not to be perceived as yet.

We have tried to discover whether a greater agreement might perhaps be obtained, if instead of the proportion of the olfaction-values those of the logarithms of the olfaction-values were taken. The subjoined tables account for this.

If in these tables the values are looked up that in the tables to the right and left are about the same, it soon appears that no manner of regularity can be observed.

Hereupon Mr. HERMANIDES has tried a vectorial representation of

PROPORTIONAL NUMBERS OF THE LOGARITHMS OF STIMULI COMPENSATING EACH OTHER

The Table gives the value of $\frac{log\ p}{log\ q}$.

ZWAARDEMAKER.

log q \ log p	I	II	III	IV	V	VI	VII	VIII	IX
I		1.25	0.85	12.5	2.27	∞	5.88	0.67	5.26
II	0.8		0.86	1.35	1.79	1.15	2.5	0.78	2.44
III	1.18	1.16		2.27	2.38	1	3.57	1.32	1.69
IV	0 08	0.74	0.44		1.01	0	0.7	0.94	1.54
V	0.44	0.56	0.42	0.99		0.33	0.96	0.8	1.41
VI	0	0.87	1	∞	3.08		∞	3.09	0
VII	0.17	0.4	0.28	1.43	1.04	0		0.74	0.56
VIII	1.5	1.28	0.76	1.06	1.25	0.11	1.35		1.18
IX	0.19	0.41	0.49	0.65	0.71	∞	1 79	0.85	

HERMANIDES.

log q \ log p	I	II	III	IV	V	VI	VII	VIII	IX
I		1.09	0.41	2.94	1.11	0.63	2.44	0.47	1.96
II	0.92		0.67	1.09	1.64	1.19	4.54	0.68	1.35
III	2 43	1.49		3.45	1.49	1.89	15.15	1.28	2.63
IV	0.34	0.92	0.29		0.64	0.52	1.23	1.32	2.38
V	0.9	0 61	0.67	1.55		0.78	1.18	0.84	2
VI	1.58	0.84	0.53	1.91	1.28		10	0.85	2.08
VII	0.41	0.22	0.066	0.81	0.85	0.1		0.38	1.25
VIII	2.14	1.47	0.78	0.76	1 19	1 18	2 66		1.82
IX	0.51	0.74	0.38	0.42	0.50	0.48	0.8	0.55	

the results of his observations. Whereas at the time, among the 252 possible combinations, I myself had found only one system [1]) in which three odours, considered as vectors, could be joined together, HERMANIDES succeeded in finding three such systems.

The system of ZWAARDEMAKER was that of terpineol, aethylbisulfide and guajacol in the proportion 1 : 18 : 1 and as such not be construed; the systems of HERMANIDES appear to be:

A. isoamylacetate, guajacol and valerian-acid in the proportion 2 : 1 : 29.

B. nitrobenzol, aethylbisulfide and pyridine in the proportion 1.9 : 1 : 0.5.

C. guajacol, pyridine and scatol in the proportion 1.5 : 1 : 24.

On account of the relative rarity of such constellations and the want of agreement between the system of ZWAARDEMAKER and those of HERMANIDES no further inferences can, at least provisionally, be drawn from this.

An important fact, however, is that in the further work some identities between vectors for both of us conjointly have been found. The subjoined table accounts for this.

TABLE of the coinciding of two vectors with respect to two others, both for Z and for Hs.

Vectors becoming identical	Vectors whose position is given beforehand
isoamylacetate and guajacol	valerian acid and scatol
aethylbisulphide and valerian acid	guajacol and pyridine
valerian acid and scatol	isoamylacetate and terpineol
valerian acid and scatol	isoamylacetate and guajacol

Also the reciprocity between the proportion of isoamylacetate and guajacol on the one side and valerian-acid and scatol on the other, previously observed by me, has been found back by HERMANIDES, as a look at the table may show.

Likewise manifold identity. For four vectors it occurred, just as with me, only once and that for the odours:

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[1]) Through an error in the calculation, Proceedings Vol. XVI, p. 187 note, the only possible combination of three vectors in my system has been erroneously given. The unique constellation is that of Terpineol, Aethylbisulfide and Guajacol. Inferences were not drawn then, nor are they tried now.

isoamylacetate
terpineol
guajacol
pyridine

Placed as vectors over against the odours valerian acid and scatol, likewise thought as vectors.

In that constellation, as we see, we meet with pyridine, in contradistinction to nitrobenzol in my case. If we leave this element, which is different, aside and if we take into consideration only what they have in common, we are in a position, in both my odour-system and that of HERMANIDES, to put two groups of odours over against one another.

Group A.	*Group B.*
isoamylacetate	valerian acid
terpineol	scatol
guajacol	

In these two groups terpineol and guajacol, on the ground of the reciprocity mentioned just now, stand in the same proportion to each other as valerian acid and scatol.

Cases of odour-compensations, showing both for Z and Hs about the same proportions.

Odour-compensations	Z	Hs	Ha
Isoamylacetate : Guajacol	3	2	5.7
Nitrobenzol : Terpineol	1.37	2.22	28
Nitrobenzol : Muskon	0.43	0.81	0.29
Nitrobenzol : Guajacol	0.65	0.61	49
Nitrobenzol : Valerian acid	0.03	0.013	0.8
Nitrobenzol : Pyridine	3	3.99	10.0
Terpineol : Muskon	0.12	0.18	0.05
Terpineol : Valerian acid	0.050	0.062	0.026
Terpineol : Pyridine	0 50	0.58	0.093
Muskon : Aethylbisulfide	1.0	2.27	0.15
Muskon : Pyridine	1.2	0.55	0.16
Aethylbisulfide : Valerian acid	1.2	0.58	0.13
Aethylbisulfide : Pyridine	3.2	1.85	0.45
Aethylbisulfide : Scatol	0.023	0.037	0.036
Guajacol : Valerian acid	0.030	0.035	0.013

We have been in a position to cause the combinations in which we agree, to be examined by a third person, Mr. H. HERINGA, assistant at the laboratory. In order to make a comparison possible, the cases meant are put here side by side.

Of the proportional numbers in column 4 (Ha) three deviate very little from the corresponding values in column 2 (Z) and 3 (Hs), viz. terpineol to valerian acid, aethylbisulfide to scatol, guajacol to valerian acid (strictly speaking it is only aethylbisulfide to scatol that remains within the limits of the probable errors; the others are, speaking roughly, in agreement with each other).

If we connect this with the division into two ordinary groups *A* and *B*, we are led to single out from the nine standard-groups three groups, whose action upon the normal olfactory organ is presumably characteristic and in general equal for different persons (at least three observers got about the same quantitative proportions, when trying these three odours in combinations).

They are the odours:

From group A.	*From group B.*
terpineol	valerian acid
aethylbisulfide	scatol
guajacol	valerian acid

It is to be recommended for the present to continue the investigation with these three odours, for it has been these three materials, chemically sharply defined, which have exercised a quantitatively agreeing influence on the sensorial complex of three observers, mutually quite independent of each other. Therefore it may be assumed that the action of these odours, apart from the accidental psychological appreciation, within the limit of the probable errors and based upon the physiological organisation, possesses a sharply outlined nature and a very definite degree of intensity.

About the results of the bilateral combination of odours I hope I shall be able to offer a communication later on, when the three odours, which usually act in the same way, shall have been investigated on this point by more observers.

Chemistry. — *"The electromotive force of zinc amalgams"*. By Prof. ERNST COHEN and W. TOMBROCK. (Communicated by Prof. P. VAN ROMBURGH).

(Communicated in the meeting of May 29, 1909).

1. After it had been found, in the development of the thermodynamics of the normal cells [1]), that the deviations which the E. M. F. of the WESTON-cell exhibits under definite circumstances must be attributed to the peculiar behaviour of the cadmium amalgam used therein, the researches of H. C. BIJL [2]) have elucidated the matter so that we are now in a better position to judge. Yet, the latest researches of ERNST COHEN and H. R. KRUYT [3]) have proved that the last word has not yet been spoken in this matter and that a continuation of the study of the cadmium amalgams in the direction indicated will be necessary before we arrive at the construction of an accurate temperature formula for that standard [4]).

2. An investigation of the zinc amalgams whose electromotive force will have to be known accurately if the CLARK-cell is really to be used as a standard, was undertaken by one of us some years ago [5]). This field, however, is nearly quite barren. The most confused ideas still reign, as may be shown by the fact that the American Electrochemical Society at the Meeting, May 1907 made the following theme a subject of discussion "If an electrode of zinc and another of amalgamated zinc be placed in a solution of zinc and the electrodes connected externally, a current will flow in the external circuit from the zinc plate to the amalgamated one and zinc will be deposited in the former. What is the source of energy of this battery? It has been thought to be thermal, and that the liquid cools by virtue of this reaction, thereby absorbing heat from the air. If so, it would offer a direct means of changing the heat energy of the air into an electric current, and would seem to be contrary to the second law of thermodynamics."

We will again refer in detail to the theory of that phenomenon in our paper which will appear shortly in the Zeitschrift für physikalische Chemie. Here it may be pointed out that, first described by

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[1]) ERNST COHEN. Proc. 1900 Vol. III, p. 91.
[2]) Zeitschr. für physik. Chemie **41**, 641 (1902).
[3]) Zeitschr. für physik. Chemie **65**, 359 (1909).
[4]) Proc. 1900 Vol. III, p. 225.
[5]) Electrochemical and Metallurgical Industry, **5**, 93 (1907).

HUMPHRY DAVY [1]), it attracted the attention of FARADAY [2]), GAUGAIN [3]), POGGENDORFF [4]), HENRICI [5]), ANTOINE BECQUEREL [6]), REGNAULT [7]), WILLARD GIBBS [8]), ROBB [9]), GABRIEL LIPPMANN [10]) and SPENCER [11]) and that theories to explain the *polar exchange* which was supposed to occur therein have not been wanting. We may state, however, that this phenomenon does not take place if only due precautions are taken and that there is no question of a "conflict" with the second law of thermodynamics.

3. In order to exclude secondary reactions our determinations for the measurement of the E.M.F. of different concentrated amalgams were carried out at 0°.5. The details of the experiments will be described fully later on. It should be mentioned that the materials used comply with high demands as to purity.

Chemically pure zinc possesses towards the solution of its salts a sharply defined potential, but only then when it has been deposited electrolytically in dendritic, or fungoid form [12]). We replaced it by a 10% amalgam such as is used in CLARK's normal cells and constructed H-shaped cells according to the scheme:

Zinc amalgam 10% by weight	dilute solution of $ZnSO_4$	Zinc amalgam x% by weight

The x% amalgams (x between 2,5 and 6%) used therein were prepared by weighing and melting the materials with special precautions. The amalgams were then introduced into the H-shaped cells.

The amalgams of a lower concentration were investigated by means of the little apparatus shown in the Fig.

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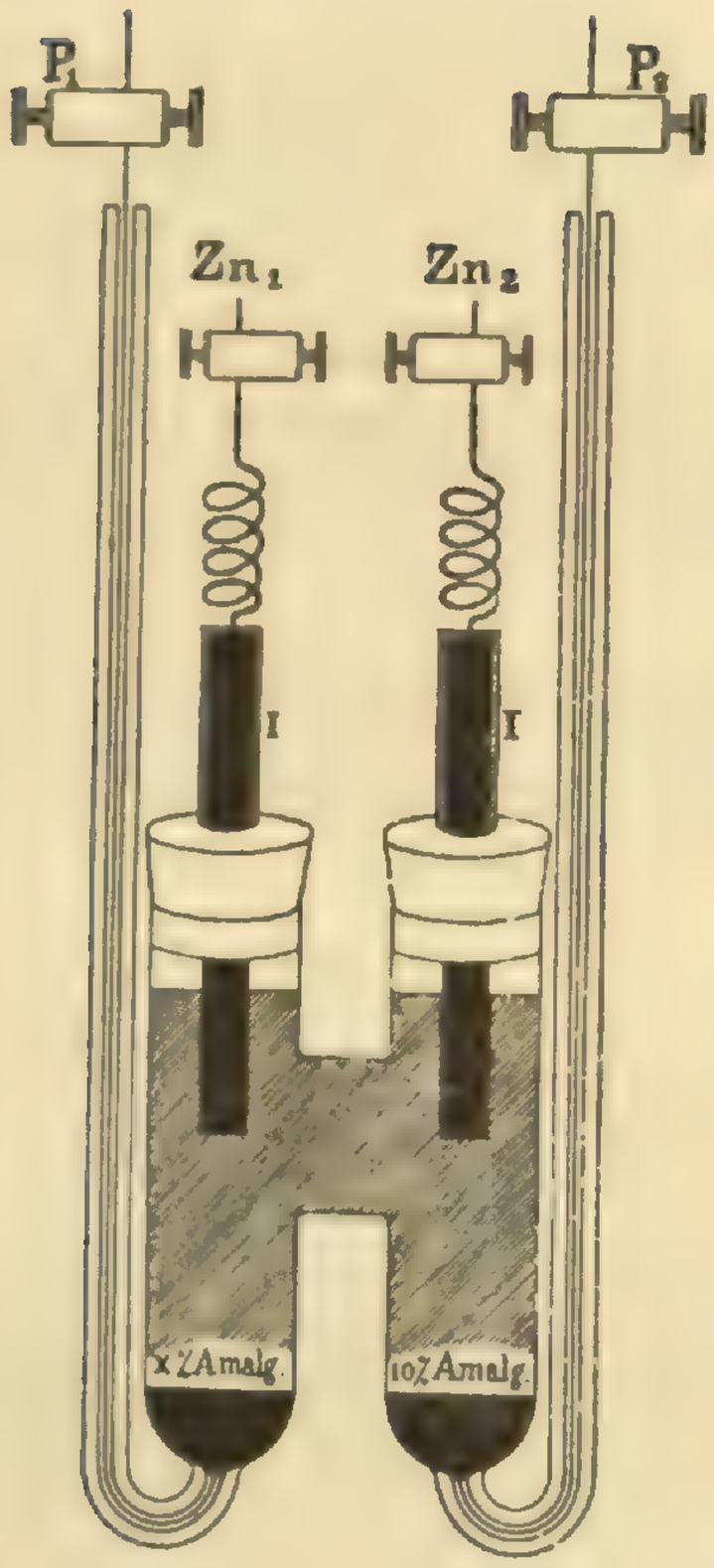
1) Phil. Trans. Roy. Soc. **116**, 405 (1826).
2) Phil. Trans. Roy. Soc. 1834, Part. I 459 § 1004 en 1005.
3) C. R. **42**, 430 (1856).
4) POGGENDORFFS Annalen **50**, 262 (1840).
5) POGGENDORFFS Annalen **58**, 376 (1843).
6) Traité d'Electricité et de Magnétisme, Paris 1854. Tome 1, p. 221.
7) Ann. de chim. et de physique (3) **44**, 453 (1855).
8) Transactions of the Connecticut Acad. of Arts and Sciences **3**, 343 (1878).
9) WIEDEMANNS Annalen, N. F. **20**, 798 (1883).
10) Journ. de Physique (2) **3**, 388 (1884).
11) Zeitschr. für Elektrochemie **11**, 681 (1905).
CROVA, Ann. de chim. et de phys. (3) **69**, 458 (1863).
HOCKIN and TAYLOR, Journ. Soc. Tel. Ing. **8**, 282 (1879).
ST. LINDECK, WIEDEMANNS Ann. N. F. **35**, 311 (1888).
12) TH. W. RICHARDS and LEWIS, Zeitschr. für physik. Chemie **28**, 1 (1899).



On the bottom of the right limb is placed a $10^0/_0$ zinc amalgam and in the left limb a weighed amount of pure mercury. The whole is filled up with a solution of zinc sulphate not saturated at 0°.5 (its reaction was perfectly neutral towards congo-red). I and I are two rods of chemically pure zinc which are in communication with the polar terminals Zn_1 and Zn_2, while P_1 and P_2 lead to the mercury and the $10^0/_0$ amalgam respectively. The apparatus is placed in a thermostat (0°.5) and Zn_1 and P_1 are now put in a circuit, in which is placed an accumulator with a resistance coil, in such a manner that on closing the same Zn_1 acts as anode and P_1 as cathode. A certain amount of metallic zinc is then precipitated from the zinc sulphate solution which dissolves in the mercury and so yields an amalgam. In that circuit we also put a milliampèremeter; so if the time which the current has passed is known, the concentration of the amalgam formed will be accurately known. Our milliampèremeter was carefully checked by means of a silver coulometer, but by way of control we also put in during the experiment two silver coulometers besides the milliampèremeter.

If, now (by means of a swing) P_1P_2 or Zn_2P_2, respectively, is introduced into the potentiometer of POGGENDORFF—DU BOIS-REYMOND, the E.M.F. between the $10^0/_0$ zinc amalgam and the x-$^0/_0$ amalgams formed may be measured at any moment, and also that between pure zinc (in rod shape) and the $10^0/_0$ amalgam.

Before each measurement the whole cell was shaken vigorously in order to stir up the zinc solution (whose concentration might change locally in consequence of the electrolysis) and also the amalgam.

5. Whereas for the sake of brevity we omit the description of these measurements and of the controlled apparatus employed we give in the following tables the results of our measurements. It should be stated that all the experiments were made in duplicate.

As required by theory the $10^0/_0$ amalgam forms at all concentrations where there is still an E. M. F. the negative (solution pole). The second and third column show how little defined is the potential of pure zinc; the rods Zn_1 and Zn_2 were two halves *of the same rod.*

TABLE I.

Temperature 0°.5 C.

E. M. F. in Volts of a 10°/₀ Zinc amalgam towards:
(This amalgam forms the *negative* pole).

Amalgam of x°/₀ Zn	Zinc pole Zn_1	Zinc pole Zn_2
x=0.103 0.0285	0.0086	0.0146
0.309 0.0161	0.0079	0.0146
0.619 0.0078	0.0073	0.0113
1.031 0.0024	0.0065	0.0097
1.134 0.0016	0.0066	0.0090
1.237 0.0007	0.0069	0.0082
1.340 0.0000	0.0064	0.0076
1.443 0.0000	0.0067	0.0075
1.856 0.0000	0.0060	0.0066
2.268 0.0000	0.0060	0.0062

This again shows that zinc in this form is unfit for potential measurements.

In the tables now following the E. M. F. of the amalgam cells with amalgams of higher concentration are indicated at 0°.5 in volts. The times are hours after the construction of the cells. The + and — signs placed before the potential difference indicate that the *least* concentrated amalgam forms the positive, or the negative pole respectively of the cell investigated.

TABLE II.

10°/₀ amalgam—2.57°/₀ amalgam.

Time	E. M. F. in Volts.	
	Cell I	Cell II
0	— 0.0003	— 0.0003
0.5	0.0000	0.0000
3.5	+ 0.0001	0.0000
11.75	+ 0.0001	+ 0.0001
22.25	— 0.0001	0.0000

TABLE III.

10% amalgam—3.40% amalgam.

Time	E. M. F. in Volts.	
	Cell III	Cell IV
0	+ 0.0004	+ 0.0001
0.5	+ 0.0002	0.0000
1.25	+ 0.0001	+ 0.0001
7.25	+ 0.0002	+ 0.0001
17.50	+ 0.0003	+ 0.0000

TABLE IV.

10% amalgam—4.34% amalgam.

Time	E. M. F. in Volts.	
	Cell V	Cell VI
0	+0.0007	0.0000
0.5	+0.0001	0.0000
21.3	0.0000	0.0000

TABLE V.

10% amalgam—7.88% amalgam.

Time	E. M. F. in Volts.	
	Cell VII	Cell VIII
0	0.0000	0.0000
21.3	0.0000	0.0000

TABLE VI.

10% amalgam—20.34% amalgam.

Time	E. M. F. in Volts.	
	Cell IX	Cell X
0	+0.0001	+0.0001
3.5	+0 0004	+0.0004
7	+0.0001	+0.0001
3.5	+0.0001	+0.0001

TABLE VII.

10% amalgam—38.82% amalgam..

Time	E. M. F. in Volts	
	Cell XI	Cell XII
0	+0.0001	0.0000
4	0.0000	0.0000
7	+0.0001	+0.0001
13.5	+0.0001	+0.0001

TABLE VIII.

10% amalgam—60% amalgam.

Time	E. M. F. in Volts	
	Cell XIII	Cell XIV
0	+ 0.0008	+ 0.0007
3.5	+ 0.0006	+ 0.0006
7	+ 0.0006	+ 0.0006
13.5	+ 0.0001	+ 0.0001
23.5	0.0000	+ 0.0008
29.0	+ 0.0001	+ 0.0010
30.5	— 0.0008	+ 0.0008
31.25	—	+ 0.0008
37.25	—	+ 0.0009
47.75	+ 0.0009	+ 0.0009

The previous measurements of ERNST COHEN have taught us that at 0° the potential difference between pure (dendritic) zinc and the 10% amalgam amounts to 0.000488 volts, the zinc forming the negative pole. From the above measurements we may conclude that in the cases where a potential difference between zinc and zinc amalgams does exist, this is always of such a nature that the zinc forms the negative pole.

All the objections arising from the fact that previous observations have been made with materials insufficiently electrically defined are thus removed and the "explanations" given for the occurrence of *polar exchange* also no longer count.

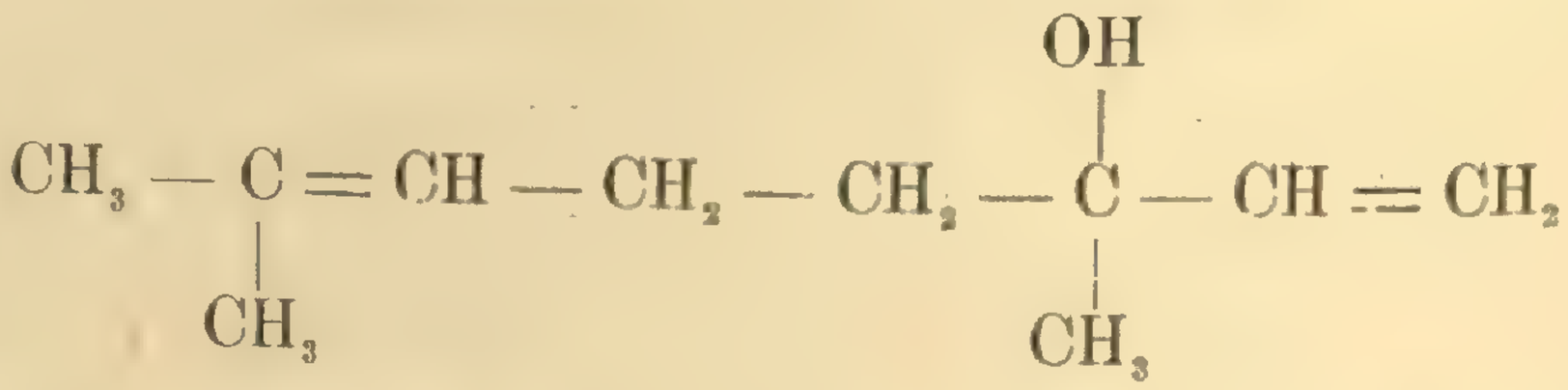
As soon as the investigation as to the relation between the progressive change of the melting point line of the zinc amalgams and their electromotive force which is being carried out by Mr. P. J. H. VAN GINNEKEN, is completely drawn to a close we will revert to the question in regard to the CLARK-cells.

Utrecht, May 1909. VAN 'T HOFF Laboratory.

Chemistry. — *"On the action of active copper on linalool."* By Dr. C. J. ENKLAAR. (Communicated by Prof. P. VAN ROMBURGH.)

(Communicated in the Meeting of May 29, 1909).

In a previous investigation[1]) I have shown, that the tertiary aliphatic terpene alcohol linanool may be converted by means of active nickel and hydrogen into a saturated tertiary alcohol and a decane; I have prepared this alcohol synthetically and thus demonstrated the position of the (OH)-group in the formula of linalool. As in consequence, the existing differences of opinion as to this structural formula have been removed, I now accept with TIEMANN and SEMMLER the following formula for linalool:



The usual methods to eliminate the elements of water from tertiary alcohols, and so to prepare hydrocarbons, are often incomplete when the products of the reaction are variable. For instance by the action of acids on linalool the terpenes dipentene and terpinene[2]) occur amongst the products formed, whose structures bear no relation to the structural formula of the starting product. We may, therefore safely assume that the formation of dipentene and terpinene is accompanied by a radical shifting of bonds either in the linalool molecule or in that of a primarily formed hydrocarbon. In my opinion, the real dehydration product of linalool has, therefore, escaped notice and final products of an "inversion" have been obtained, which nearly every other terpene would have yielded.

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[1]) Rec. Trav. Chim. **27**, 411 (1908).

[2]) BERTRAM and WAHLBAUM, J. pr. Ch. [2], **45**, 601 (1892).

The above does not altogether apply to the action of acetic anhydride on linalool, which has been studied successively by BARBIER [1]), STEPHAN [2]) and ZEITSCHEL [3]). The last investigator has obtained a hydrocarbon, whose sp. gr. ($d_{15} = 0.813$) was considerably lower than that of dipentene and terpinene and comes near to that of the aliphatic terpenes; ZEITSCHEL speaks of "ein olefinisches, dem Myrcen sehr nahe stehendes Terpen" [4]). It appeared to me desirable to attempt the dehydration of linalool with a neutral or alkaline substance.

SABATIER and SENDERENS have found in active copper (prepared by reducing copper oxide at 200°) a contact substance, which accelerates addition of hydrogen in a less degree than active nickel, but which can split up tertiary alcohols into water and hydrocarbons. A specimen of linalool of high optical rotation ($\alpha_D = -17°14'$), kindly presented to me by Dr. GILDEMEISTER, chief-chemist to SCHIMMEL and Co., was passed at 135° over active copper and fully one-half was split up into hydrocarbon and water. By fractionating the product of the reaction, finally over metallic sodium, the main product, an optically active hydrocarbon of the formula $C_{10}H_{16}$ is obtained. The constants of the fractions are:

Fraction	B.p.$_{12}$	d_{15}	α_D (100 mM)	$n_{d_{11}}$	$H_\gamma - H_\alpha$
I	66°—68°	0,8401			(mol. disp.)
II	67.5°—68.5°	0,8344	+ 8° 8'		
III	68.5°—69°	0,8339		1,4802	1.84
IV	69°—70°	0,8340		(MR = 46,1)	

The boiling point of the first fraction at the ordinary pressure is 171°—173°. The odour of the three last fractions reminds one of that of limolene, that of the first one of myrcene. This fraction also differs in the constants whereas the other fractions agree. Assuming that, in the main, a single substance is obtained, which according to the constants cannot be limonene or terpinene (these terpenes have a 8° higher b. p.), I have more closely investigated the fractions II—IV and now give a preliminary communication of the results of the investigation.

The molecular refraction and dispersion are very peculiar. Of the

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[1]) Compt. rend. **116**, 1200 (1893).

[2]) J. pr. Ch. [2], **58**, 109 (1898).

[3]) Ber. **39**, 1780 (1906).

[4]) Compare: STEPHAN, pr. Ch. [2] **58**, 113 (1899) and SEMMLER, Aetherische Oele, Bnd I, 549 (1906).

values found 46,1 and 1,84 the refraction lies between the one calculated for a monocyclic terpene (45,2) and that of an aliphatic terpene (46.9); the dispersion exceeds that of a monocyclic terpene by 28.6% and even that of an aliphatic terpene by 11.5%. I have noticed something similar [1]) with aliphatic terpenes with a conjugated system of double bonds. A similar configuration might be suspected in the structure of the substances obtained and something might be said as to the presence of a monocyclic terpene with two double bonds in a conjugated position.

I have also investigated the action of bromine, hydrogen and potassium permanganate. The bromine absorption was carried on in carbon tetrachloride solution; one mol. rapidly absorbed two atoms of bromine, but (as shown by the decoloration) a third atom was absorbed but very slowly with evolution of hydrogen bromide. The bromide obtained was a colourless oil. The hydrogen absorption was carried on by passing the substance very slowly at 180° over active nickel in a current of hydrogen. The product of the reaction may be separated into three parts by fractionation, namely a first fraction with a low sp. gr. corresponding in odour with dimethyl-2-6-octane [2]), a second (main) fraction boiling at 164°– 166° ($d_{15} = 0{,}787$), of the composition $C_{10}H_{18}$ and a third fraction boiling at 166° – 170°. Nearly the whole of the liquid passed over below 169°, the b. p. of hexahydro-p-cymene, which ought to have formed if a monocyclic terpene derivative of p-menthane had been present.

The result of this reaction gives some insight into the nature of the products. SABATIER and SENDERENS were able to effect the hydrogenation of bicyclic terpenes to hydrocarbons of the composition $C_{10}H_{18}$, whilst the monocyclic terpenes investigated all yielded the same hexahydro-p-cymene [3]). In addition, I have also investigated a terpene with a 6- and 3-cycle in the molecule, namely, sabinene which also yielded a hydrocarbon $C_{10}H_{18}$. The said hydrocarbons $C_{10}H_{18}$ are all different, as different cycle-systems occur in their molecules. In all of them I have found a characteristic property; they are saturated towards potassium permanganate, but are rapidly attacked by bromine at the ordinary temperature. This reaction is shown in a very characteristic manner by dihydrosabinene. In the above mentioned hydrogenation product of the hydrocarbon from linanool, I also noticed this property from which it ought to be concluded that a bicyclic terpene occurs in the original product; if this is a single

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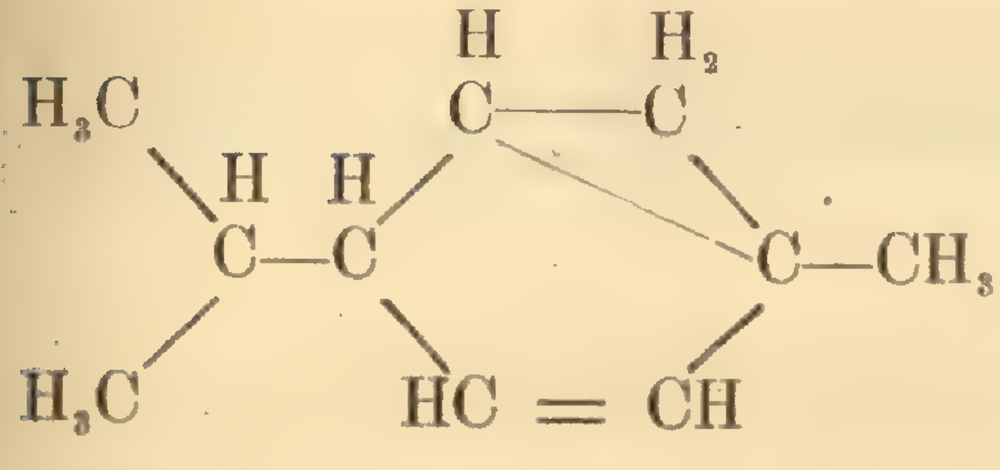
[1]) Rec. Trav. Chim. **27**, 432 (1908).
[2]) C. J. ENKLAAR, Ber. **41**, 2083 (1908).
[3]) Ann. de Chim. et de Phys. [8]. IV, 1905 p. 319.

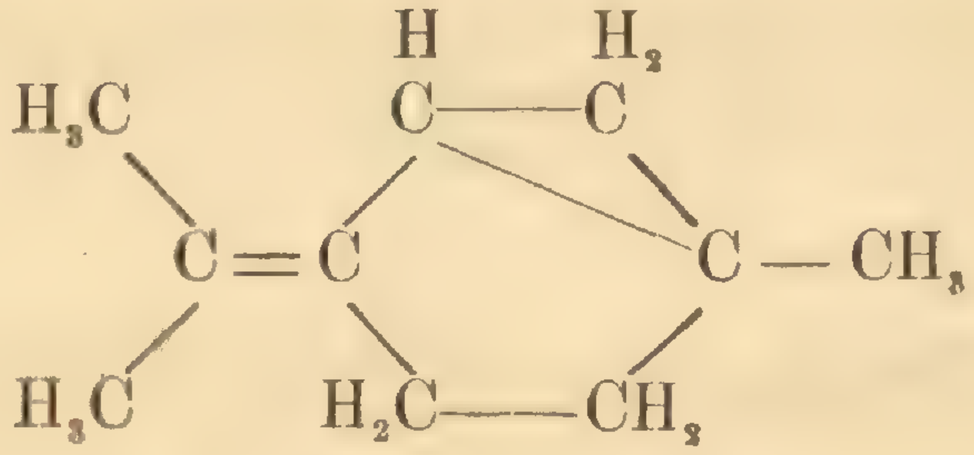
substance it must consist entirely of the same. For the present, however, I will abstain from stating the probable quantity thereof, but we may say that the rather high optical activity of the product supports this view and we might think of a substance of one of the following formulae.

I



or

II



The high refraction and dispersion may be due to the presence of a trimethylene cycle in conjunction with a double bond. Both the action of bromine and of hydrogen pointed, therefore, to the presence of a bicyclic terpene. Potassium permanganate at first forms a glycol the composition of which is probably $C_{10}H_{18}O_2$. On oxidation or by the action of dilute acids, its aqueous solution becomes turbid with separation of an oil having a characteristic odour. By the action of potassium permanganate are formed also considerable quantities of acids, among which a butyric acid and a non-volatile acid, which on oxidation with hydrogen peroxide passes into an oxy-acid. Of the original hydrocarbon it may also be mentioned that it absorbs oxygen and gives with nitrous acid an oily nitrite. I cannot quite reject the idea that the hydrocarbon might be phellandrene mixed with a substance of low sp.gr. Moreover its constants agree very closely with a terpene which SEMMLER [1]) has obtained from sabinene and with the β-terpinene of WALLACH. SEMMLER considers his terpene as a representative of a new class of terpenes, which possess in their molecule a 5-cycle and two double bonds; a further research will follow.

By the action of active copper, linalool is, therefore, dehydrated with closing of the ring. I also found another method in which the closing of the ring is avoided as much as possible. It consists in heating the alcohol (1 mol.) with phenylisocyanate (2 mols.) at 140°—150°. A constant current of carbon dioxide is evolved while s-diphenylurea crystallises out. By distillation a hydrocarbon can be obtained of low sp. gr. (0,810). In odour it resembles myrcene and like this it may be reduced by sodium and alcohol to a hydrocarbon

[1]) Ber. **39**, 4414 (1906)

of the boiling point of dihydromyrcene; with a mixture of glacial acetic and sulphuric acids it gives the characteristic odour of myrcenol. I am engaged in a closer investigation of this product and in working out the method.

The continuation and extension of this provisional research has been rendered possible to me by the support of the provincial Utrecht Society of Arts and Sciences. The optical determinations have been carried out in the organic-chemical laboratory at Utrecht.

's-Hertogenbosch, May 1909.

Botany. — "*A brief contribution to the knowledge of endozoic seed distribution by birds in Java, based on a collection made by Mr.* BARTHELS, *on the Pangerango and near Batavia.*" I. By S. H. KOORDERS.

(*Contribution to the knowledge of the Flora of Java.* V).

(Communicated in the meeting of May 29, 1909).

§ 1. General remarks and method.

Mr. M. BARTHELS of Tjisaat, Preanger, Western Java, who has such an intimate knowledge of the javanese bird-fauna, was kind enough to comply with my request for a collection of seeds and fruits from the stomachs and intestines of birds, shot by him on the Pangerango and near Batavia. This collection, though small, had been made with great care; it was accurately labelled and was preserved in alcohol.

For this collection, which I received here (at Leiden) from India last May, the scientific names of the birds had already been established by Mr. BARTHELS, but the botanical determination of the fruits and seeds had not yet been made. The botanical determination has now been accomplished, as far as time and the nature of the material would permit, the material of 's Rijks Herbarium at Leiden having been used for comparison.

In each case I have included in quotation-marks, adding the letters M. B. between parentheses, those data which were given by Mr. BARTHELS on the collecting labels, relating to the name of the birds, the place and date of collection and the serial number of his ornithological collection.

The ornithological investigations of the late Dr. VORDERMAN, of

Mr. M. Barthels and especially also of Dr. J. C. Koningsberger [1]) have supplied many data, particularly as regards the names of insects found in the stomachs of javanese birds and the animal food of "useful" and "harmful" birds; on the other hand our knowledge of the significance of various species of fruit-eating birds for the distribution of seeds and fruits in Java is still extremely limited.

At any rate, methodical observations on this question are almost completely wanting in the literature for Java (with numerical data about the number and botanical origin of the seeds and the scientific name of the birds). For only two Javanese plant species [*Lantana Camara* L., *Myrica javanica* Bl.] special data are to be found in the literature [2]).

I have therefore attempted to ascertain as well as possible at least the natural order and the genus of the seeds, collected by Mr. Barthels.

Thanks to the method of labelling employed, the further determination of the species can take place later, when more time and material are available. I have contented myself in most cases with the probable determination of the plant-genera.

Meanwhile germination experiments, made on the spot, remain very desirable for the future. In such experiments one portion of the seeds found in birds stomachs, will have to be sown at once, while the other portion is preserved with the seedlings for botanical determination. For the collection, which has now been examined, such comparative germination experiments are still wanting, because Mr. Barthels could not afford the necessary time for them.

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[1]) Koningsberger, J. C., Aanteekeningen over maaginhouden van vogels (in Teijsmannia XIX. (1908) p. 83—99).

[2]) Ernst, A., Besiedelung vulkanischen Bodens auf Java, etc. (in Karsten & Schenck, Vegetationsbilder. I. Reihe, Heft 1 u. 2. (1909) IV. Text von Tafel 9 und 10).

Koorders, S. H., Contribution N°. 1 to the knowledge of the flora of Java § 1. On the oecological conditions, means of dissemination and geographical distribution of the species of Myricaceae, growing wild in Java, especially in the higher mountains (in Kon. Akademie v. Wetenschappen in Amsterdam, Verslag der Gew. Vergadering d. Wis- en Natuurk. Afd. 12 Maart 1908. p. 645—652 and the literature quoted there; also the English translation in: Kon. Akademie v. Wetenschappen Amsterdam. Proceeding 24 April 1908 p. 674—680.

Valeton, Th., Distribution of fruits by animals. (Dutch) in Teijsmannia. IV. (1881) p. 205—222. (*Lantana Camara* L., p. 217).

Cordes, H. J. W. The djatiwoods of Java. 1881. (Batavia) p. 81—83 (Dutch). This deals with the distribution of the fruits of a shrub (*Lantana Camara* Linn.). which was introduced into Java from S. America about 1850 and now covers many thousands of acres in Java. See also the literature mentioned there: Natuurk. Tijdschr. Ned. Indië XXXI. p. 287 and Tijdschrift voor Nijverheid en Landbouw Ned. Indië XXII. p. 75.

For a large number of European plants Dr. SELIM BIRGER has recently made and published numerous feeding experiments [1]). For Java however, germination experiments with seeds and fruits from birds stomachs have, as far as I know, only been made with *Myrica javanica* Bl. (compare footnote 2 above on p. 109).

In order to obtain a better insight into the significance of endozoic seed-dissemination by birds in the Dutch East Indies and especially in Java, special comparative feeding-experiments are further very desirable with some species of birds, which are the most important from a phyto-geographical point of view. With these experiments on captive birds it would also be desirable to choose in each case a portion of the seeds and fruits from *common, wild plants* and always to keep a portion of the seeds in order to check the botanical determination. By combining the serial numbers (of the birds) with the letters *a, b, c,* for the seeds and fruits, the origin of each of the seeds can thus be sharply defined scientifically, and a confusion with other seeds and fruits be excluded.

As the first example of this simple method of numbering, which has been found to serve its purpose well, I choose here the following combination: BARTHELS n. 5903*a* means for instance the seeds and fruits of the plant species *a* found in the stomach of the bird Coll. BARTHELS n. 5903 on 18 X 1908 in Western Java on the Pangerango. And in this case (see below) the bird n. 5903 was, according to the original label, determined by Mr. BARTHELS as ♂ Criniger gularis Horsf., where as the vegetable stomach contents *a* has been provisionally determined by me as Micromelum pubescens Bl.

As a second example the following combination may serve: BARTHELS n. 6162*a* and BARTHELS a. 6162*b*. Here n. 6162*a* therefore signifies (see below) the seeds with portions of the fruit wall of the species of *Euphorbiaceae* while the combination 6162*b* indicates the fruit-stones of the species of *Myrsinaceae,* both of which were found in the stomach contents of the bird, shot by Mr. BARTHELS on 30 I. 1909 in Western Java on the Pangerango and determined by him as ♀ *Poliomyias luteola* Pall. (winter visitor!).

With the aid of this combination of a number and a letter, the provisional botanical determination can always be confirmed or corrected with ease.

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[1]) Compare e. g. SELIM BIRGER, Über die Pflanzen mit endozoischer Verbreitung (in Botaniska Notizer 1907). — Abstract by ULBRICH in Literaturbericht in ENGLER's Botan. Jahrb. Bd. 40 (1907) p. 215. — SELIM BIRGER, Über endozoische Samenverbreitung durch Vögel (in Särtvijck Svensk Botanisk Tidskrift. Bd. I. (1907) 1—31 and abstract by ULBRICH in ENGLER's Botan. Jahrb. Bd. 41 (1908) Heft. 3. p. 68.

§ 2. List of Javanese birds with the botanical origin of the seeds and fruits found in their stomach contents.

"Aleippa solitaria *Cab.* ♂. — 12. X. 1908. — Pangerango. — Coll. BARTHELS Nr. 5892" (M. B.).

Stomach contents (5892*a*): 4 fruits and 12 undamaged seeds of one species of Araliaceae, probably of the genus Schefflera Forst.

"Carpophaga lacerunulata *Temm.* ♂. — 20. IX. 1908. — Pangerango. — Coll. BARTHELS Nr. 5837". (M. B.).

Stomach contents 5837*a*: one undamaged seed, still surrounded by its arillus, of the order Myristicaceae and presumably of Knema laurina Bl. or of Knema glauca Bl.

"Chloropsis nigricollis *Veill* ♂. — 8. II. 1909. — Pangerango. — Coll. M. BARTHELS". Nr. 6180. (M. B.).

Stomach contents 6180*a*: Two ripe fruits of a species of Loranthaceae and presumably either of the genus Loranthus *Linn.* or of the genus Elytranthe *Blume*. Further in the same intestines: six ripe fruits of the same species of Loranthaceae.

"Chloropsis nigricollis *Viell.* ♂. — 30. I. 1909. — Pangerango. — Coll. M. BARTHELS Nr. 6161. (M. B.).

Stomach contents 6161*a*: Three seeds of a Dicotyledonous species, not as yet further determined.

"Chloropsis nigricollis *Viell.* ♀. — 12. X. 1908. — Pangerango. — Coll. M. BARTHELS. Nr. 5891". (M. B.).

Stomach contents 5891*a*: too much damaged; could not be determined.

"Cochoa azurea *Temm*; ♂. — 5. XI. 1908. — Pangerango. — Coll. M. BARTHELS. Nr. 5940". (M. B.).

Stomach contents 5940*a*: Seven undamaged ripe fruits of one species of Rutaceae, probably of the genus Fagara *Linn.*

"Criniger gularis *Horsf.* ♂. — 15. X. 1908. — Pangerango. — Coll. M. BARTHELS". Nr. 5897. (M. B.)

Stomach contents 5897*a*: three undamaged ripe seeds with fragments of the fruit pulp of one and the same species of Araliaceae

and very probably of Arthrophyllum diversifolium Bl.

"**Criniger gularis** *Horsf.* ♀. — 2. X. 1908. — Pangerango. — Coll. M. Barthels. Nr. 5870". (M. B.).

Stomach contents 5870*a*: Four undamaged ripe well developed seeds, and some young rudimentary ones, in addition to pieces of the fruit valves and wall of one and the same species of Euphorbiaceae and probably of the genus Glochidion.

„**Criniger gularis** *Horsf.* ♂ — 18. X. 1908. — Pangerango. — Coll. M. Barthels. Nr. 5903". (M. B.).

Stomach contents 5903*a*: Two undamaged ripe seeds and some fragments of the fruit pulp of one species of Rutaceae, and presumably of Micromelum pubescens Bl.

"**Cyanops armillaris** *Temm.* ♂. — 21. I. 1909. — Pangerango. — Coll. M. Barthels Nr. 6143". (M. B.).

Stomach contents 6143*a*: Ten undamaged ripe seeds of one species of Euphorbiaceae, probably of the genus Antidesma and perhaps of the arboraceous Antidesma tetrandum Bl. which is commonly wild on the Pangerango.

"**Cyanops armillaris** *Temm.* ♂. — 25. VII. 1908. — Coll. M. Barthels. Nr. 5645". (M. B.).

Stomach contents 5645*a*: Fragments of ripe receptacula of one species of Moraceae namely of Ficus.

"**Hemiscus virescens** *Temm.* ♂. — 17. X. 1908. — Pangerango. Coll. M. Barthels. Nr. 5902". (M. B.).

Stomach contents 5902*a*: Fragments of ripe receptacula of a species of Moraceae, of the genus Ficus.

"**Hemiscus virescens** *Temm.* ♀. — 20. XI. 1908. — Pangerango. — Coll. M. Barthels. Nr. 5973". (M. B.).

Stomach contents 5973*a*: eighteen ripe undamaged seeds with fragments of the values of dehisced fruits of one species of the order Euphorbiaceae, presumably of the genus Glochidion.

"**Laniellus leucogrammicus** *Swains.* ♂. — 19. VII. 1908. — Pangerango. — Coll. M. Barthels. Nr. 5634". (M. B.).

Stomach contents 5634*a*: Fragments of ripe fig fruits of one species of Ficus (order Moraceae).

"**Limonites ruficollis** *Pall.* ♀. — 18. XII. 1908. — Batavia. — Coll. M. Barthels. Nr. 6051. (Wintervisitor!)". (M. B.).

Stomach contents 6051*a*: seven ripe, partially damaged fruits of one species of Cyperaceae, presumably of the genus Carex, the fruit resembling that of Carex baccans Nees., but differing from it.

"**Poliomyias luteola** *Pall.* ♀. — 30. I. 1909. — Pangerango. — Coll. M. Barthels. (Wintervisitor!). Nr. 6162". (M. B.).

Stomach contents 6162*a*: seven undamaged ripe seeds with fragments of the walls of septa of the fruits of one species of Euphorbiaceae, presumably of the genus Glochidion.

Stomach contents 6162*b* (in the same stomach): three ripe stones of one species of Myrsinaceae and presumably of Embelia Ribes *Burm.* or of a closely related species.

"**Pycnonotus bimaculatus** *Horsf.* ♀. — 26. VII. 1900. — Pangerango. — Coll. M. Barthels. Nr. 5646". (M. B.).

Stomach contents 5646*a*: Five undamaged ripe fruits of one species of Myrsinaceae and presumably of Embelia Ribes *Burm.*

"**Pycnonotus bimaculatus** *Horsf.* ♀. — 14. XI. 09. — Pangerango. — Coll. M. Barthels. Nr. 5958". (M. B.).

Stomach contents 5958*a*: Twenty-four ripe seeds of one species of the order Euphorbiaceae, presumably belonging to the genus Glochidion.

§ 3. Brief survey, arranged alphabetically, according to plant orders.

Araliaceae: Arthrophyllum diversifolium Bl. = Nr. 5897*a* (Criniger); Schefflera spec. = 5892*a* (Aleippa).

Cyperaceae: Carex? spec. = Nr. 6051*a* (Limonites).

Euphorbiaceae: Antidesma spec. = Nr. 6143*a* (Cyanops). Glochidian spec. = Nr. 5870*a* (Criniger); Nr. 5958*a* (Pycnonotus); Nr. 5973*a* (*Hemiscus*) and Nr. 6162*a* (Poliomyias).

Loranthaceae: Elytranthe or Loranthus spec. = Nr. 6180*a* (Chloropsis).

Moraceae: Ficus spec. div. = Nr. 5643*a* (Laniellus); Nr. 5645*a* (Cyanops) and Nr. 5902*a* (Hemiscus).

Myristicaceae: Knema laurina? Bl. = Nr. 5837*a* (Carpophaga).

Myrsinaceae: Embelia Ribes Burm. = Nr. 5646*a* (Pycnonotus).
Rutaceae: Fagara spec. = Nr. 5940*a* (Cochoa); Micromelum pubescens Bl. = Nr. 5903*a* (Criniger).

List of the birds' stomachs numbers and the generic names determined by Mr. M. BARTHELS of the birds of the collection BARTHELS: Nr. 5634 (Laniellus); 5645 (Cyanops); 5646 (Pycnonotus); 5837 (Carpophaga); 5870 (Criniger); 5891 (Chloropsis); 5892 (Aleippa); 5897 (Criniger); 5903 (Criniger); 5940 (Cochoa); 5902 (Hemiscus); (Pycnonotus); 5973 (Hemiscus); 6051 (Limonites); 6143 (Cyanops); 6161 (Chloropsis); 6162 (Poliomyias); 6180 (Chloropsis).

§ 4. Phytogeographical remarks on the occurrence of the enumerated plant-genera in the district where the birds were shot.

Araliaceae. — Arthrophyllum pinnatum BL. is a small tree, bearing fruit profusely. (See stomach contents Nr. 5897*a*, Criniger). It is very common in the woods on the lower slopes of the Pangerango and also elsewhere in Java, especially in young secondary woods at an altitude of 0—1500 m.

Of the genus Schefflera *Forst.* there occur wild in Java 14 more or less arborescent, or merely fruticose species. Some of these species occur wild in the lower and higher woods of the Pangerango and one arborescent species (Schefflera rugosa Harms) occurs very commonly in the highest forests on the summits of the Pangerango at 2500—3030 m. above sea-level.

Some species of Schefflera not only occur in the moist mountain forests of the Pangerango as soil plants, but are also found high up in the crowns of the forest-giants as epiphytes (Comp. Nr. 5892*a*, Aleippa).

Cyperaceae. The genus Carex is represented in Java by some species, especially in the higher cooler mountain regions and for instance also in the Pangerango. A few species (e. g. Carex baccans Nees., C. filicina Nees., C. Rafflesiana Boot. and C. hypsophila Miq.) are, for instance, common at 2500—3030 m. above sea level on the highest summit of the Gede and the Pangerango. In the lower regions Carex is rare in Java (comp. stomach contents Nr. 6051*a*, Limonites).

Euphorbiaceae. The genus Glochidion *Forst.* is represented in Java by a number of arborescent wild species, some of which

are very abundant e. g. in the lower mountain woods of the Pangerango. In the highest forests in the summit of the Pangerango at an altitude of 3000 m. the genus Glochidion is completely wanting. (Comp. stomach contents Nr. 5870*a*, Criniger, etc.).

Of the genus Antidesma there occur in Java, in addition to the much cultivated *A. Bunias* Linn., a number of tree like wild species especially in the lower mountain forests. A very common small forest-tree, bearing profuse fruit, and occurring in the mountain forests of the Pangerango is e. g. Antidesma tetrandrum Bl. (Comp. stomach contents Nr. 6143*a*, Cyanops).

Loranthaceae. The genera Loranthus and Elytranthe are represented in Western Java, among others also in the Pangerango by Loranthus lepidotus Bl., which at an altitude of 0—2500 m. occurs very commonly as a parasite on the most widely different species of trees, and further by Elytranthe globosa (*Roxb.*) *Engler* and E. avenia (*Bl.*) *Engler*. (See stomach contents Nr. 6180*a*, Chloropsis).

Moraceae. The genus Ficus is represented in Java by more than 80 wild-growing species of which some are trees, some are shrubs and a few are climbing-plants.

A large number of species live in their youth for a time epithytically. Numerous Ficus species occur in the mountain-forests of the Pangerango and a few species ascend to the forests on the highest summit.

Among the very common, richly fruiting arborescent species of the lower Pangerango forests, the following may be mentioned, which occur also elsewhere in Java: *F. toxicaria* Linn., *Ficus Ribes Reinw.* and *Ficus variegata Bl.*

Myristicaceae. — Knema laurina Bl. and Knema glauca Bl. are two species of nutmeg trees which occur also in the forests on the lower slopes of the Pangerango. They generally do not ascend in Western Java higher than 1200 M. above sea-level. (Compare stomach contents Nr. 5857*a* Carpophaga).

Myrsinaceae. — Embelia Ribes *Burm.* is a woody climber, of frequent occurrence in the whole of Western Java and for instance also in the lower woods of the Pangerango; it bears fruit very copiously. (See stomach contents Nr. 5646*a*, Pycnonotus).

Rutaceae. — Of the genus Fagara the species F. scandens (*Bl.*) *Engler* occurs not infrequently in Western Java, for instance on the Pangerango in the lower mountain forests: it is a high-climbing, woody species bearing copious fruit and ascending to about 1300 M. above sea-level. (Compare stomach contents Nr. 5940*a*, Cochoa).

Micromelum pubescens Bl. is a small tree, which is extremely frequent in the whole of Java from the plain to the lower mountain forests, and also pretty abundant on the Pangerango, for instance, up to 1200 M. in young, secondary woods. It nearly always bears plentiful fruit frequently. (Compare stomach contents Nr. 5903*a*, Criniger).

In conclusion it is my very pleasant duty to offer my cordial thanks to Mr. M. Barthels of Tjisaat, Preanger, for sending me the seeds etc. from birds' stomachs and for determining the bird species, from which the above mentioned material was collected by him.

All the botanical determinations and the above text, in as far as it has not been placed between quotation-marks (see above) are due to myself.

Leiden, May 25, 1909.

Botany. — "*Some remarks on the nomenclature and synonymy of Xylosma leprosipes Clos., X. fragrans Decne and Flueggea serrata Miq.*" By S. H. Koorders. (*Contribution to the knowledge of the Flora of Java.* VI.)

(Communicated in the meeting of May 29, 1909).

§ 1. On the authentic specimens of Xylosma leprosipes Clos. and X. fragrans Decne.

I have received from Paris a fragment of the authentic herbarium specimen which was collected about 1805 by Leschenault de la Tour in *Java* (probably in the Rahoen-Idjen mountains) and which was described in 1857 by Prof. D. Clos as a new species under the name of Xylosma leprosipes Clos. The receipt of this specimen has enabled me to ascertain, that it is quite identical with a javanese species, described by Miquel in 1859 (Fl. Ind. Bat. I. 2. p. 105) as the type of a new genus under the name Bennettia

Horsfieldii Miq.. I was able to examine authentic herbarium material of the latter species in 's-Rijks Herbaria at Leiden and at Utrecht.

As a result of this identity, and following the rules for nomenclature of the Vienna botanical congress, the name B. Horsfieldii of Miquel cannot be maintained and must give way to Bennettia leprosipes (Clos.) Koord., nomen nov.

The above-mentioned fragment of an authentic specimen of Xylosma leprosipes Clos. was presented to me some years ago at my request by Prof. Bois of the Museum d' Histoire Naturelle, together with a fragment of the authentic specimen of Xylosma fragrans Clos.

It was only recently, however, that I discovered that Xylosma leprosipes Clos. has been erroneously regarded in the literature as a species of the genus Xylosma G. Forst, and must be transferred from this genus to the genus Bennettia Miquel, described by Miquel l.c. in 1859; at the same time I found the only javanese species of this genus to be quite identical with Xylosma leprosipes Clos.

Xylosma fragrans Decaisne is however, according to the authentic specimen, very sharply marked off from X. leprosipes Clos, and has been correctly placed by Decaisne in the genus Xylosma G. Forst, which is synonymous with Myroxylon J. & G. Forst.

The latter species (X. fragrans) was, according to the above-mentioned material, collected in Timor. This locality, which was written on the label of the fragment in Prof. Bois' autograph, agrees well with the specimens of Xylosma fragrans Decaisne which I found in 's Rijks Herbarium of Leiden. There this species is only represented by specimens from Timor. These were collected in that island by Spanoghe, Decaisne, Brown and Zippelius.

In the literature and in 's Rijks Herbarium X. fragrans is therefore only known from Timor, and hence not yet from Java, for instance. I found however in 's Rijks Herbarium a duplicate of a sterile undetermined specimen of a very rare species of tree, collected by me in Java (Kds n. 10019 β from Herb. Buitenzorg), which closely resembled some of the Timor specimens. The sterile Javanese specimen (Kds n. 10019 β) is, however, distinguished by the following characteristics: foliis junioribus laxe sericeis, glabrescentibus; innovationibus dense sericeis.

Since the flowers and fruits of this javanese tree are still wanting and the original specimen of the Buitenzorg Herbarium is not at my disposal now, it does not seem desirable to decide the question whether

or no it belongs to Xylosma fragrans Decne (sensu latiore).

Bennettia leprosipes, (Clos) Koord. nom. nov.; Bennettia Horsfieldii, Miq. ! Fl. Ind. Bat. I. 2. (1859) 105; Xylosma leprosipes, Clos! in Ann. Sc. Nat. Sér. 4. VIII. (1857) 233.

Hitherto this species had only been mentioned in the literature for Java (comp. Koord. & Valeton Bijdr. Booms. Java V. (1900) p. 30; Warburg and Engler & Prantl Natürl. Pflanzenfam. III. 6*a*. p. 45). I have now, however, also found in 's Rijks Herbarium at Leiden specimens from Sumatra, which are beyond any doubt identical with the Javanese Bennettia leprosipes. These specimens are: Sumatra, Padangsche Bovenlanden, on the Singalan (Beccari ! nr. 51 and 324, anno 1878), Sumatra, Hochangkola, Batak countries, in the woods at 300—1000 metres above sea-level (Junghuhn ! — anno 1839? = Herb. Lugd. Bat. n. 127 (898/260)).

The javanese specimens of Bennettia leprosipes in 's Rijks Herbarium at Leiden are the following: Java, withour further indication of locality (Horsfield. — ex. Herb. Utrecht). Java, without further locality (Graf von Hoffmannseg ex Herb. Hasskarl). Western Java, Preanger (Plantae Junghuhn. ! ineditae n. 546, 547, 548 and 549). — Central-Java, G. Oengaran (Junghuhn !). I did not observe in 's Rijks Herbarium any specimen from Eastern Java, although this species was also found by me in that part of Java in very large numbers, e.g. in the woods of the Rahoen-Idjen-mountains at 1000—1500 metres above sea level; it was also collected by me. (comp. Koord. & Valeton l. c. 30). The species is frequent in the whole of Java below 1600 metres above sea level, in heterogeneous, evergreen woods.

It is my pleasant duty to record here my thanks to Prof. D. G. J. M. Bois of Paris, for his kindness in sending me the authentic specimens of *Xylosma fragrans* Decaisne and of *Xylosma leprosipes* Clos.

Xylosma amara, (Spanoghe) Koord. nomen nov.; X. fragrans, Decaisne ! in Voy. Venus. Botan. (1835) tab. 25 absque diagn.; Clos. in Ann. Sc. Nat. Sér. 4. VIII. (1875) 232; Flacourtia amara Spanoghe in Linnaea XV (1841) 166; Myroxylon amara, (Span.) Warburg u. Engler u. Pratl Natürl. Pflanzenf. III. 6*b* (1894) 41: Rhamnus timoriensis, Zipp. ! msc. in Herb. Lugd. Batav.

Timor (Zippel.; Spanoghe; Decaisne; R. Brown). —? Java.

§ 2. On the authentic specimen of Flueggea serrata Miq.

In the herbarium revision of the wild-growing Euphorbiaceae of the higher mountains of Java I have found in the Herbarium of the University of Utrecht the authentic specimen which served

MIQUEL for his diagnosis of Flueggea? serrata MIQ. MIQUEL's diagnosis of this species, which is published in his Flora Ind. Bat. I. 2. (1859) p. 356, is not quite sufficient for clear recognition.

A closer examination of this authentic specimen showed that MIQUEL had very properly queried the generic name Flueggia, for my examination gave the surprising result, that the species was not only incorrectly placed by MIQUEL in this Euphorbiaceous genus, but that the specimen does not even belong to the order Euphorbiaceae, but to the order Celastraceae, and not to a new species, but to a species of the genus Celastrus LINN., namely to Celastrus paniculata WILLD. (Spec. Pl. I. p. 1125). This species has already been described under many names, is very variable and widely distributed and consists of a number of elementary species. According to the original label, this herbariumspecimen Utrecht was collected by JUNGHUHN in the alpine regions of Central Java; it is merely a particularly small-leaved specimen of a species of Celastrus, the small size of the leaves being presumably due to unfavourable alpine oecological conditions. The shrub occurs in various parts of Java, up to an altitude of 2000 m. and generally does not climb high.

In the Kew Herbarium and also in 's-Rijks Herbarium at Leiden I have been able to compare the authentic specimen, of the Utrecht Herbarium, referred to above, (Flueggea? serrata MIQ.) with very numerous specimens of Celastrus paniculata WILLD. from widely different localities, and in so doing I have also found a few small-leaved specimens of this species of Celastrus which closely agree with it.

The synonymy of the species thus becomes as follows:

Celastrus paniculata, WILLD. (sensu latiore) Spec. Pl. I. 1125; MIQ. Fl. Ind. Bat. I. 2. (1859) p. 590; HOOK. Fl. Br. India I. (1875) 617; C. alnifolia, DON. Rodr. 190; C. dependens, WALL. Catal. (1825) n. 4302; C. multiflora, ROXB. Fl. Ind. I. 622; C. nutans, ROXB. Fl. Ind. I. 623; C. Rothiana, ROEM. & SCHULT. Syst. V. 423; C. Metziana, TURCZ. in Bull. Soc. Nat. Mosc. (1858) 447; Ceanothus paniculatus, ROTH. Nov. Spec. 154; Scutia paniculata, DON. Gen. Syst. II. 34; Flueggia serrata, MIQ.! msc. in MIQ. Plant. Jungh. ined. msc.; MIQ.! Fl. Ind. Bat. I. 2. (1859) 356.

I wish to express my thanks to the Directors of the Herbaria of Kew, Leiden, and Utrecht for the opportunity given me of examining the material in question.

Leiden, Mei 25, 1909.

Physics. — *"On the solid state."* III. By J. J. VAN LAAR. (Communicated by Prof. H. A. LORENTZ).

(Communicated in the meeting of May 29, 1909).

10. Before proceeding to the discussion of the course of the p-T-line liquid-solid, we shall first adduce some more material to prove our statements. For the values assumed by us of a, b, etc. (Comp. II p. 27) we shall namely successively calculate, besides the already calculated isotherm for $T = 9$ (see I and II), also the isotherms of 100°, 128°, 144°, 160°, 200° and 400° (all absolute temperatures), in order to show that on increase of temperature they really present the shapes and transitions indicated by us. (see the Plate of Part I, fig. 1—3).

The accurate course of all these isotherms $p = f(v)$ for $T = 0$, 9, 100, etc. has been indicated on the Plate belonging to this paper. In view of the available dimensions the scale of the pressure-values had to be somewhat reduced, which appears particularly for the critical isotherm ($T = 400$), where $p_c = 400$, i.e. $400 \times 41{,}2 = 16480$ atms. — or after division by 100 (see II, p. 27 [1])) $= 165$ atms., which is rendered comparatively small in the drawing. The values of v have not been indicated further than $v = 3$ (300), so that the last maximum no longer falls inside the limits of the drawing.

The isotherm $T = 0$ (comp. II, p. 34) is indicated by a line of alternate dashes and dots [2]). Further the locus of the maxima D and of the minima C, which passes through the point of inflection I, where C and D coincide, has been indicated by an ordinary dotted line. We may remind the reader that the solid phase (for so far as it is realizable) always lies on the portion CD of the isotherms, the liquid

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[1]) So all the pressure-values of the following tables must still be multiplied by 0,412; all the values of volume by 100.

[2]) The values of p in the curvilinear part ED have been calculated from the equation $p = -\frac{q_0}{-\Delta b} - \frac{a}{v^2} = 6400 - \frac{a}{v^2}$ [see II, p. 31, formule (8), which apparently applies to all the points between E and D, where $\varphi = \frac{1}{\theta} = \infty$, so that $(p + a/v^2)(-\Delta b) = q_0$ (see II, p. 31-32)]. The corresponding values of β may be calculated for this part from the relation $v = b$ holding there $\left(\text{for } \varphi = \frac{1+\beta}{v-b}(-\Delta b) = \infty\right)$, so that β becomes $= \frac{b_1 - v}{-\Delta b}$. (See form. (11) of II, p. 35).

phase on the portion EF. The vapour phase (between A and B) lies outside the limits of our drawing.

The locus of the minima in E and of the maxima in B, which passes through the critical point K, where B and E coincide, has not been indicated in our drawing.

Nor are the pressures of coexistence liquid-solid, solid-vapour, liquid-vapour indicated by horizontal lines; only for $T=0$ the pressure of coexistence liquid-solid is indicated ($p=1000$), whereas that for solid-vapour coincides with the axis $p=0$.

$$\underline{T=100.}$$

Now with $\theta=\frac{200}{3200}=\frac{1}{16}$ the formula (3^a) [see II, p. 26] becomes:

$$\frac{\beta^2}{1-\beta^2}=32000\times\left(\frac{1}{16}\right)^{3/2}e^{-16}\frac{e^{\varphi}}{\varphi}=500\ e^{-16}\frac{e^{\varphi}}{\varphi},$$

hence

$$log^{10}\frac{1-\beta^2}{\beta^2}=-4{,}250+0{,}4343\,\varphi-log^{10}\varphi,$$

while for p we find $p=400\,\varphi-\frac{2700}{v^2}$ (see II, p. 28).

$\varphi = \frac{1+\beta}{v-b}(-\Delta b)$	$log^{10}\ \frac{\beta^2}{1+\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
23	4,377	$2{,}38 \times 10^4$	$1 - \frac{1}{48000}$	0,54	+ 60
17	1,903	80,0	0,994	0,56	—1760
16	1,495	31,3	0,98	0,57	—1920
15	1,089	12,3	0,96	0,58	—1900
13	0,282	1,91	0,81	0,66	— 920
12	—0,117	0,763	0,66	0,74	— 130
11	—0,514	0 306	0,48	0,83	+ 440
10	—0,907	0,124	0,33	0,90	+ 670
9,5	—1,102	0,0791	0,27	0,93	+ 600
9	—1,293	0,0509	0,22	0,96	+ 660
7	—2,055	0,00881	0,093	1,03	+ 260
5	—2,777	0,00167	0,041	1,08	— 300
3	—3,424	0,000265	0,016	1,16	— 800
2	—3,682	0,000208	0,014	1,25	— 940
1	—3,815	0,000153	0,012(min.)	1,50	— 800
0,5	—3,732	0,000185	0,014	2,0	— 470
0,25	—3,539	0,000289	0,017	3,0	— 190
0,1	—3,206	0,000622	0,025	6,1	— 32
0,03	—2,726	0 00188	0,043	18	+ 4,0
0,02	—2,542	0,00287	0,054	27	4,4
0,01	—2,245	0,00568	0,075	55	3,1
10^{-3}	—1,249	0,0563	0,23	620	0,39
10^{-4}	—0,250	0,563	0,60	8000	0,040
10^{-5}	0,750	5,63	0,92	96000	0,004
10^{-6}	1,750	56,3	0,991	10^6	0,0004
0	∞	∞	1	∞	0

The minimum value of β for $v = 1{,}5$ ($\varphi = 1$), which amounted only to 10^{-38} for $T = 9$, has now already risen to $0{,}012 = 10^{-2}$.

From the approximate relation $\frac{v^3}{(v-1)^2} = \frac{2a}{RT} = 27$, which holds for B and C if $\beta = 0$ (see II, p. 33), we should find $v_C = 1{,}28$ and $v_B = 24{,}9$. And as $v = 1 + \frac{1}{2\varphi}$ for $\beta = 0$, we have $\varphi_C = 1{,}8$ and $\varphi_B = 0{,}021$, while then the corresponding values of p are further $p_C = -930$, $p_B = 4{,}0$. So these approximate values agree pretty well with the exact values of the table.

$T = 128$.

Now with $\theta = \frac{256}{3200} = \frac{1}{12{,}5}$ (3^a) becomes:

$$\frac{\beta^2}{1-\beta^2} = 32000 \times \left(\frac{1}{12{,}5}\right)^{3/2} e^{-12{,}5}\, \frac{e}{\varphi},$$

or

$$log^{10} \frac{\beta^2}{1-\beta^2} = -2{,}569 + 0{,}4343\, \varphi - log^{10} \varphi,$$

whereas further $p = 512\, \varphi \quad \frac{2700}{v^2}$.

φ	$log^{10}\frac{\beta^2}{1-\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
17	3,584	3830	$1-\frac{1}{7700}$	0,56	+ 61
12	1,563	36,6	0,987	0,59	— 1630
11	1,167	14,7	0,97	0,61	— 1730
10	0,774	5,94	0,93	0,63	— 1600
9	0,386	2,43	0,84	0,68	— 1210
8	0,002	1,01	0,71	0,75	— 660
7	— 0,374	0,423	0,55	0,84	— 260
6	— 0,741	0,181	0,39	0,92	— 120
5	— 1,096	0,0801	0,27	0,99	— 190
4	— 1,434	0,0268	0,19	1,05	— 380
3	— 1,743	0,0181	0,13	1,12	— 610
2	— 2,001	0,00997	0,099	1,23	— 770
1	— 2,135	0,00733	0,085(min.)	1,5	— 690
0,5	— 2,051	0,00890	0,094	2,0	— 390
0,25	— 1,858	0,0139	0,12	3,2	— 140
0,1	— 1,525	0,0290	0,17	6,8	— 7,8
0,05	— 1,246	0,0567	0,23	13	+ 10,1
0,04	— 1,154	0,0702	0,26	17	+ 10,7
0,03	— 1,033	0,0927	0,29	22	+ 10,0
0,01	— 0,565	0,273	0,46	74	4,6
0,001	0,432	2,70	0,85	930	0,51
0,0001	1,431	27,0	0,98	9900	0,051
0	∞	∞	1	∞	0

The minimum value of β is almost 0,1 at this temperature.

In the maximum at D the pressure is now negative, so that we are already above the triple-point temperature at 128°.

$T = 144.$

With $\theta = \frac{288}{3200} = \frac{9}{100}$ the equation (3^a) becomes:

$$\frac{\beta^2}{1-\beta^2} = 32000 \times \left(\frac{9}{100}\right)^{3/2} e^{-\frac{100}{9}} \frac{e^{\varphi}}{\varphi},$$

hence

$$log^{10}\frac{\beta^2}{1-\beta^2} = -1{,}889 + 0{,}4343\,\varphi - log^{10}\,\varphi,$$

while p is calculated from $p = 576\,\varphi - \frac{2700}{v^2}$.

φ	$log^{10}\frac{\beta^2}{1-\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
14,5	3,149	2810	$1-\frac{1}{5600}$	0,57	+ 16
10	1,454	28,4	0,98	0,61	— 1550
9	1,065	11,6	0,96	0,63	— 1640
8	0,682	4,81	0,91	0,66	— 1510
7	0,306	2,02	0,82	0,72	— 1160
6	—0,061	0,868	0,68	0,80	— 770
5	—0,416	0,383	0,53	0,89	— 530
4	—0,754	0,176	0,39	0,98	— 510
3	—1,063	0,0865	0,28	1,07	— 620
2,5	—1,201	0,0629	0,24	1,13	— 690
2	—1,321	0,0477	0,21	1,20	— 733
1,5	—1,414	0,0386	0,19	1,30	— 730
1	—1,455	0,0351	0,18 (min)	1,5	— 620
0,5	—1,371	0,0426	0,20	2,1	— 320
0,25	—1,178	0,0663	0,25	3,4	— 90
0,10	—0,846	0,143	0,35	7,6	+ 10,7
0,07	—0,731	0,186	0,40	10,8	+ 17,3
0,06	—0,641	0,229	0,43	12,7	+ 17,9
0,05	—0,566	0,271	0,46	15,4	+ 17,4
0,01	0,115	1,30	0,75	89	5,4
0,001	1,111	12,9	0,96	980	0,57
0,0001	2,111	129	0,996	10000	0,058
0	∞	∞	1	∞	∞

Again β_{min} is larger, now already $= 0{,}18$. The maximum at D and the minimum at C have drawn very near to each other; we shall presently see, that at 160° the coinciding takes place in

a point of inflection $I = C,D$. (Cf. Fig. 3 of the plate of part I).

$$\underline{T = 160.}$$

With $\theta = \frac{320}{3200} = \frac{1}{10}$ we have now:

$$\frac{\beta^2}{1-\beta^2} = 32000 \times (1/10)^{3/2}\, e^{-10}\, \frac{e^{\varphi}}{\varphi},$$

or

$$log^{10}\, \frac{\beta^2}{1-\beta^2} = -\,1{,}338 + 0{,}4343\,\varphi - log^{10}\,\varphi.$$

The pressure is given by $p = 640\,\varphi - \frac{2700}{v^2}$.

φ	$log^{10}\,\frac{\beta^2}{1-\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
12,5	2,994	986	$1 - \frac{1}{2000}$	0,58	— 19
8	1,233	17,1	0,97	0,64	— 1530
7	0,857	7,20	0,94	0,67	— 1536
6	0,490	3,09	0,87	0,72	— 1350
5	0,135	1,36	0,76	0,80	— 1060
4	— 0,203	0,627	0,62	0,89	— 832
3,5	— 0,362	0,435	0,55	0,95	— 775
3	— 0,512	0,308	0,49	1,005	— 753
2,5	— 0,650	0,224	0,43	1,07	— 751 (C,D)
2	— 0,770	0,170	0,38	1,15	— 745
1,5	— 0,862	0,137	0,35	1,28	— 700
1	— 0,904	0,125	0,33 (min.)	1,5	— 560
0,5	— 0,820	0,151	0,36	2,2	— 250
0,25	— 0,627	0,236	0,44	3,7	— 40
0,1	— 0,294	0,508	0,58	8,6	27,6
0,09	— 0,253	0,559	0,60	9,6	28,2
0,08	— 0,206	0,622	0,62	10,8	28,1
0,07	— 0,152	0,704	0,64	12,4	27,3
0,01	0,667	4,64	0,91	96	6,1
0,001	1,663	46,0	0,99	1000	0,64
0	∞	∞	1	∞	0

The minimum value of β is $= 0{,}33$. The horizontal point of inflection $I = C,D$ lies evidently in the neighbourhood of $\varphi = 2{,}5$, $v = 1{,}07$, $p = -750$.

At 200° not a trace is left of the horizontal point of inflection, and only the minimum E and the maximum B of the ordinary isotherm of VAN DER WAALS is to be seen, as will appear from a following table.

11. We shall, however, first give a survey of the situation of the maximum D and the minimum C at the different temperatures from $T = 0$ to $T = 160$ calculated by us.

	$T=0$	9	100	128	144	160
φ_D	∞	173	9,5	5,9	4,4	2,5
φ_C	∞	8	1,8	1,8	1,8	
β_D	0	0,027	0,27	0,37	0,45	0,43
β_C	0	$1{,}77.10^{-37}$	0,014	0,096	0,20	
v_D	1	0,99	0,93	0,93	0,94	1,07
v_C	1	1,06	1,28	1,26	1,24	
p_D	+ 3700	+ 3470	+ 690	— 120	— 500	— 750
p_C	— 2700	— 2100	— 940	— 770	— 730	

As was said before, the locus of maxima and minima (one continuous curve, on which the point of inflection is also found) is indicated by a dotted line on the plate.

If we wish to know the data of the point of inflection accurately, we may calculate them from the formulae which I drew up in the before cited paper in the Arch. Teyler for a *critical* point (i. e. for *every* point where a maximum and a minimum in the isotherm $p = f(v)$ coincide to a horizontal point of inflection), viz. (see loc. cit. p. 29 and 31):

$$\frac{v_c}{b_c} = 3\,\frac{m^2}{3m^2-n^2} \quad ; \qquad RT_c = \frac{8}{27}\,\frac{a}{b_c}\,\frac{1}{1+\beta}\cdot\frac{n^2(3m^2-2n)}{m^5}, \quad . \quad (14)$$

in which m and n have the following signification:

$$\left.\begin{aligned} m &= 1 + {}^1/_2\,\beta(1-\beta)\,(1-\varphi)^2 \\ n &= 1 + {}^3/_4\,\beta(1-\beta)\,(1-\varphi) + {}^1/_8\,\beta(1-\beta)\,(1-3\beta^2)\,(1-\varphi)^2 \end{aligned}\right\}. \quad (15)$$

Then the pressure is further determined by the equation of state, whereas β depends on T' and φ, i. e. on T' and v, according to formula (3) in II.

As $\varphi_c = \frac{(-\Delta b)(1+\beta_c)}{v_c - b_c} = \frac{{}^1/_2 (1+\beta_c)}{v_c - b_c}$, and $v_c - b_c$, as follows from the above equation for v_c, is evidently $= 2b \times \frac{n}{3m^2 - 2n}$, we have also:

$$\varphi_c = \frac{1}{4} \frac{1+\beta_c}{1 - {}^1/_2 \beta_c} \cdot \frac{3m^2 - 2n}{n} \quad , \quad . \quad . \quad . \quad . \quad . \quad . \quad (14^a)$$

because $b = b_1 + \beta \Delta b = 1 - {}^1/_2 \beta$. Further with $a = 2700$ and $R = 2$ becomes:

$$T_c = \frac{400}{(1+\beta_c)(1 - {}^1/_2 \beta_c)} \frac{n^2(3m^2 - 2n)}{m^5} \quad . \quad . \quad . \quad . \quad (14^b)$$

We further remark (see also Teyler loc. cit.) that for $\beta = 0$ or 1 the formulae (14) duly pass into $v_c = 3b_c$ and $RT_c = \frac{8}{27} \frac{a}{b_c}$, as then m and n are $= 1$. It is true that for $\beta = 1$ we find the expression $\frac{8}{27} \frac{a}{b_e} \cdot \frac{1}{2}$ for RT_c, but as all the quantities refer to double molecular quantities, $a = 4a_2$, $b_c = 2b_2$ for $\beta = 1$, hence again $RT_c = \frac{8}{27} \frac{a_2}{b_2}$.

Now in order to calculate the exact values of φ_c, β_c, and T_c from (14^a) and (14^b) in connection with (3), we may begin with assigning to φ an arbitrary value, which lies in the neighbourhood of the expected value of φ_c. With some values of β, which lie likewise in the neighbourhood of β_c, we may then calculate the corresponding values of m and n, and further those of φ_c and T_c from (14^a) and (14^b). Then we may determine β by interpolation in such a way, that the calculated value of φ corresponds with the assumed value. Then a value of T_c also belongs to this (to be found by interpolation).

Now we examine (from the above tables) what value of β corresponds with the assumed value of φ for the value of T_c determined just now (formula (3) in II). In this we shall, of course, have to interpolate again. The value of β thus found will not at once correspond with the value of β determined just now; we then repeat the whole calculation with another value of φ, till the two values of β correspond.

Thus we find e. g. with $\varphi = 2{,}5$:

		m	n	f	T_c	f'	φ
$\varphi = 2{,}5$	$\beta = 0{,}4$	1,27	0,6774	0,4838	172,8	5,144	2,251
	$\beta = 0{,}5$	1,281	0,6924	0,4916	174,8	5,114	2,557

in which for the sake of simplicity the expression $\frac{n^2(3m^2-2n)}{m^5}$ is represented by f, and $\frac{3m^2-2n}{n}$ by f'.

We see at once that we must assume $\beta = 0{,}48$ to find $\varphi = 2{,}5$, in correspondence with the assumed value. Then T_c would be $= 174{,}4$. But according to the table for 160° and that of 200° on p. 130, a value [1]) for $\beta = 0{,}59$ corresponds to this with $\varphi = 2{,}5$ which does not agree at all with $\beta = 0{,}48$. Hence the assumed value $\varphi = 2{,}5$ is not correct.

Let us now assume $\varphi = 2{,}6$.

		m	n	f	T_c	f'	φ
$\varphi = 2{,}6$	$\beta = 0{,}4$	1,307	0,6481	0,4214	150,5	5,911	2,586
	$\beta = 0{,}5$	1,32	0,6680	0,4332	154,0	5,823	2,912

So to find $\varphi = 2{,}6$, β must be $= 0{,}404$, to which corresponds $T_c = 150{,}7$. But as according to the tables $\beta = 0{,}34$ corresponds with this, also $\varphi = 2{,}6$ is not correct.

As for $\varphi = 2{,}5$ the first found value of β is 0,11 units lower than the second, whereas for $\varphi = 2{,}6$ the first is 0,064 higher, the accurate value of φ will lie in the neighbourhood of 2,55.

		m	n	f	T_c	f'	φ
$\varphi = 2{,}55$	$\beta = 0{,}43$	1,294	0,6643	0,4492	160,5	5,567	2,535
	$\beta = 0{,}44$	1,296	0,6654	0,4491	159,9	5,573	2,572

So if we take $\beta = 0{,}434$, we find $\varphi = 2{,}55$. Then T_c becomes $= 160{,}3$. To this corresponds $\beta = 0{,}439$, so that $\varphi = 2{,}55$ is still slightly too small.

		m	n	f	T_c	f'	φ
$\varphi = 2{,}56$	$\beta = 0{,}40$	1.292	0,6600	0,4462	159,4	5,586	2,444
	$\beta = 0{,}45$	1,303	0,6643	0,4437	157,9	5,645	2,640

From this follows $\beta = 0{,}430$, $T_c = 158{,}5$. This latter value, however, gives $\beta = 0{,}421$, so that $\varphi = 2{,}56$ is slightly too high.

The accurate value of φ_c is therefore $= 2{,}55 + \frac{5}{5+9} \times 0{,}01 = 2{,}554$,

to which belongs $\underline{\beta_c = 0{,}433}$, $\underline{T_c = 159{,}6}$.

For v_c we find then further the value $\underline{1{,}064}$ from

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[1]) It appears viz. from the two tables, in connection with that for 144°, that β in the neighbourhood of $\varphi = 2$ or 3 and $T = 160$ increases by about 0,011 with every degree the temperature rises.

$v = 1 - {}^1/_2\left(\beta - \frac{1+\beta}{\varphi}\right)$, whereas $p_c = 1630 - 2385 = \underline{-755}$ is found for p_c from $p = 638{,}4\,\varphi - \frac{2700}{v^2}$.

So the values found theoretically agree perfectly with the values of the point of inflection derived from the table for 160°.

12. Now the tables for $T = 200$ and $T = 400$ may follow.

$\underline{T = 200}$.

With $\theta = \frac{400}{3200} = \frac{1}{8}$ the formula for β becomes:

$$\frac{\beta^2}{1-\beta^2} = 32000 \times ({}^1/_8)^{3/2}\, e^{-8}\, \frac{e^{\varphi}}{\varphi},$$

i.e.

$$log^{10}\frac{\beta^2}{1-\beta^2} = -\,0{,}3238 + 0{,}4343\,\varphi - log^{10}\,\varphi,$$

the pressure being given by $p = 800\,\varphi - \frac{2700}{v^2}$.

φ	$log^{10}\frac{\beta^2}{1-\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
9	2,631	427	$1-\frac{1}{860}$	0,61	— 16
6	1,504	31,9	0,98	0,67	—1150
5	1,149	14,1	0,97	0,71	—1300
4	0,811	6,48	0,93	0,78	—1280
3	0,502	3,18	0,87	0,88	—1120
2	0,244	1,75	0,80	1,05	— 850
$1^1/_2$	0,152	1,42	0,77	1,21	— 660
1	0,110	1,29	0,75 (min)	1,5	— 400
0,5	0,194	1,56	0,78	2,4	— 72
0,25	0,387	2,44	0,84	4,3	+ 51
0,15	0,565	3,67	0,89	6,8	+ 62
0,10	0,720	5,24	0,92	10	+ 54
0,01	1,681	47,9	0,990	100	+ 7,7
0,001	2,677	475	$1-\frac{1}{950}$	1000	0,80
0	∞	∞	1	∞	0

The influence of the temperature on the values of β becomes stronger and stronger. Though β_{min} was only 0,33 at 160°, this value has now risen to 0,75. As we already remarked above, not a trace of a point of inflection is now found; only the minimum at E and the maximum at B of VAN DER WAALS' ordinary isotherm have been left.

$$\underline{T = 400.}$$

For β we have in this case the relation

$$\frac{\beta^2}{1-\beta^2} = 32000 \times (^1/_4)^{3/2}\, e^{-4}\, \frac{e^{\varphi}}{\varphi} = 4000\, e^{-4}\, \frac{e^{\varphi}}{\varphi},$$

as $\theta = \frac{800}{3200} = \frac{1}{4}$. So this yields:

$$log^{10}\, \frac{\beta^2}{1-\beta^2} = 1{,}865 + 0{,}4343\, \varphi - log^{10}\, \varphi.$$

Then the value of p is further found from $p = 1600\varphi - \frac{2700}{v^2}$.

φ	$log^{10}\, \frac{\beta^2}{1-\beta^2}$	$\frac{\beta^2}{1-\beta^2}$	β	v	p
∞	∞	∞	1	0,50	∞
5	3,337	2175	$1 - \frac{1}{4400}$	0,70	2490
4	3,000	1000	$1 - \frac{1}{2000}$	0,75	1600
3	2,691	491	$1 - \frac{1}{980}$	0,83	910
2	2,433	271	$1 - \frac{1}{540}$	1,00	500
1	2,299	199	0,9975 (min)	1,5	400 (K)
0,5	2,383	242	$1 - \frac{1}{490}$	2,5	370
0,25	2,576	368	$1 - \frac{1}{740}$	4,5	270
0,1	2,908	810	$1 - \frac{1}{1600}$	10	140
0,01	3,869	7400	$1 - \frac{1}{15000}$	100	16
0,001	4,865	73300	$1 - \frac{1}{150000}$	1000	1,6
0	∞	∞	1	∞	0

The temperature is here already so high, that the dissociation of the double molecules is almost complete; even for $\varphi = 1$ the minimum value is still 0,9975. If β were exactly $= 1$, the isotherm of 400° would be the *critical* one (see the calculation in the previous paper); now it is *practically* identical with it. The critical pressure is 400, the critical volume $= 1{,}5$, i.e. just three times the volume of the simple molecules ($= 0{,}5$). That the critical point is just found at the minimum value of β, is a mere chance. For in general when $\beta = 1$, v_c is $= 3 \times 2b_2$. On the other hand the minimum of β is found at $v_m = 2(b_1 - b_2) = b_1 - \Delta b$ (see before), so $v_c = v_m$, if $6b_2 = 2(b_1 - b_2)$, i.e. $b_1 = 2 \times 2b_2$. Now we assumed for our arbitrary substance $b_1 = 1$, $2b_2 = 0{,}5$, so that this condition happens to be fulfilled.

It appears from the table and the plate, that there the former minimum at E and the maximum at B have coincided in a *horizontal point of inflection*, i.e. in the *critical* point K. For temperatures higher than 400° this point of inflection, too, will gradually disappear.

The different minima E and maxima B lie all on a curve, which passes through the critical point K, because there the maximum and minimum mentioned coincide. This locus is not indicated on the plate (see p. 121)

The following table gives a survey of the situation of these maxima and minima.

	$T=0$	9	100	128	144	160	200	400
φ_E	∞	186^+	16	11	9	7	5	1
φ_B	0	$0{,}0017^+$	0,02	0,04	0,06	0,085	0,15	
β_E	1	$1 - \frac{1}{600}$	0,98	0,97	0,96	0,94	0,97	$1 - \frac{1}{400}$
β_B	0	$2{,}2.10^{-37}$	0,054	0,26	0,43	0,61	0,89	
v_E	0,50	0,506	0,57	0,61	0,63	0,67	0,71	1,5
v_B	∞	298	27	17	13	10	7	
p_E	—4400	—3840	—1920	—1730	—1640	—1540	—1300	+400
p_B	0	+0,030	+4,4	+11	+18	+28	+62	

In a fourth continuation the discussion of the general course of the p,T-line liquid-solid (the line SM in Fig. 4 of I) will follow. As to the termination point of this line at $T = 0$ (absolute), this was already fully discussed in II, § 8 (p. 31—36).

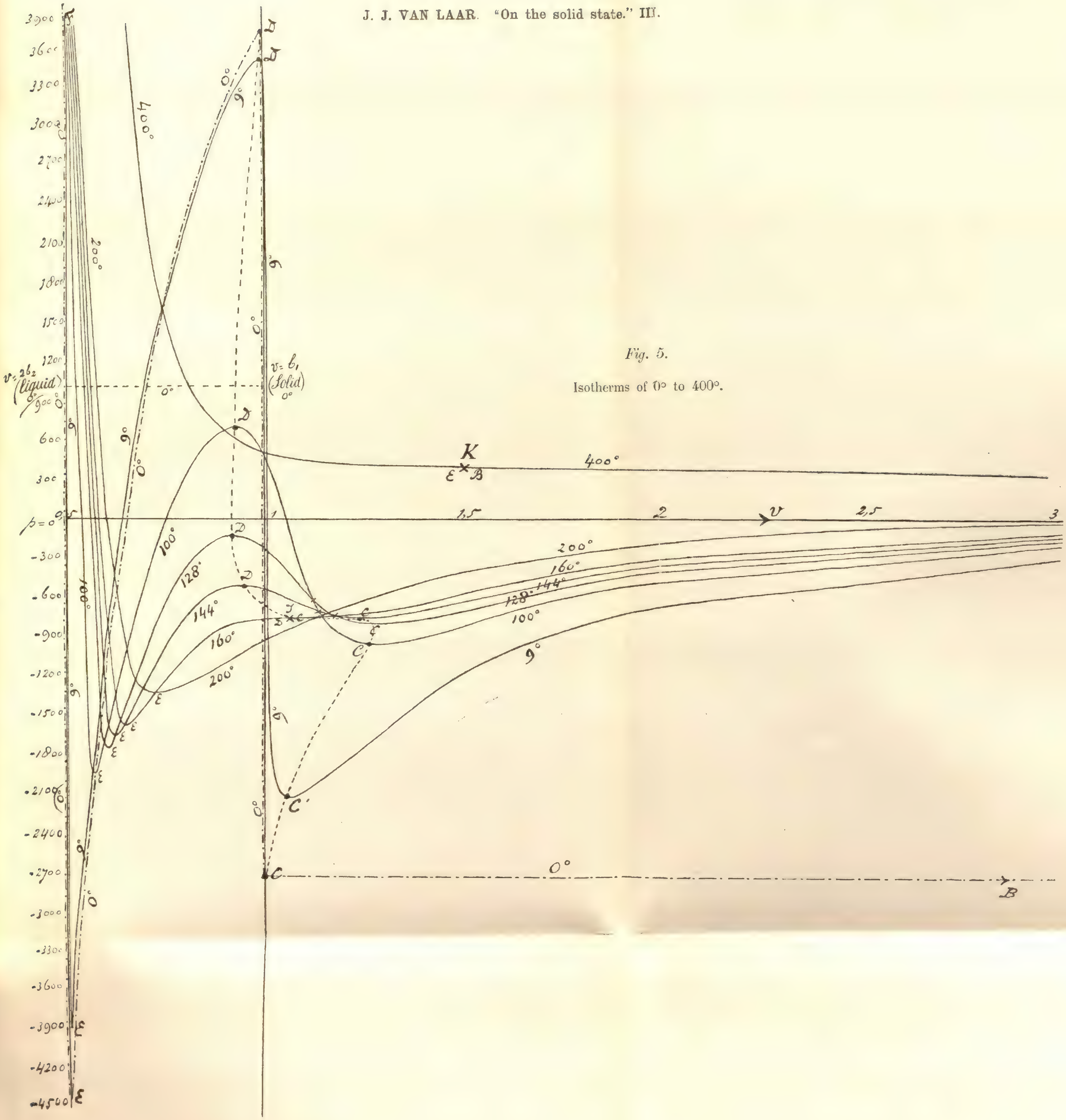

Fig. 5.

Isotherms of 0° to 400°.

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13. In the paper in the Arch. Teyler already cited a few times, I derived in § 6 (p. 34—35) the formula for the pressure of coexistence of two arbitrary phases (liquid-vapour, liquid-solid), when *association* is assumed in both phases.

I will just state the derivation of this fundamental formula here once more.

For equilibrium between two coexisting phases the molecular thermodynamic potentials of the components must be the same in the two phases. If these components are distinguished by the indices 1 (complex molecules) and 2 (simple molecules), we have e.g. in our case

$$(\mu_1)_{liq} = (\mu_1)_{solid} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (a)$$

On account of the relation of equilibrium $\mu_1 = 2\,\mu_2$ in both phases naturally $(\mu_2)_{liq} = (\mu_2)_{solid}$. Now

$$\mu_1 = C_1 - \frac{\partial \Omega'}{\partial n_1} + RT' \log c_1, \quad . \quad . \quad . \quad . \quad . \quad . \quad (b)$$

in which (see I, p. 767 and 768)

$$\Omega' = \int p dv - pv - RT\, \Sigma n_1 \,.\, log\, \Sigma n_1,$$

or

$$\Omega' = \Sigma n_1 \,.\, RT \log \frac{v-b}{\Sigma n_1} + \frac{a}{v} - pv, \quad . \quad . \quad . \quad . \quad . \quad . \quad (c)$$

when we make use of VAN DER WAALS' equation of state:

$$p = \frac{\Sigma n_1 \,.\, RT}{v-b} - \frac{a}{v^2}.$$

So we find for $\frac{\partial \Omega'}{\partial n_1}$:

$$\frac{\partial \Omega'}{\partial n_1} = RT \log \frac{v-b}{\Sigma n_1} - RT - \frac{\Sigma n_1 \,.\, RT}{v-b} b_1 + \frac{2a_1}{v},$$

because $\frac{\partial b}{\partial n_1} = b_1$ follows from $b = n_1 b_1 + n_2 b_2$, while from $a = n_1^2 a_1 + 2\, n_1 n_2 a_{12} + n_2^2 a_2$ is found $\frac{\partial a}{\partial n_1} = 2n_1 a_1 + 2n_2 a_{12} = = (2n_1 + n_2) a_1$, while a_{12} is evidently $= {}^1/_2 a_1$ (cf. also I, p. 769). Further $2n_1 + n_2 = 2(1-\beta) + 2\beta = 2$, hence finally $\frac{\partial a}{\partial n_1} = 2a_1$.

The terms with $\frac{\partial v}{\partial n_1}$ all neutralize each other in consequence of the equation of state.

We can now write for the expression found:

$$\frac{\partial \Omega'}{\partial n_1} = RT \log \frac{v-b}{\Sigma n_1} - \frac{RT}{v-b}\left[v + n_2 (b_1 - b_2)\right] + \frac{2a_1}{v},$$

because $(v-b)+\Sigma n_1 . b_1 = v - (n_1 b_1 + n_2 b_2) + (n_1 + n_2) b_1 = v + n_2 (b_1 - b_2)$.

If we still substitute $\frac{p + {}^{a}/_{v^2}}{\Sigma n_1}$ for $\frac{RT}{v-b}$, we get:

$$\frac{\partial \Omega'}{\partial n_1} = RT \log \frac{v-b}{\Sigma n_1} - \frac{p + {}^{a}/_{v^2}}{\Sigma n_1} v + \frac{2a_1}{v} - \frac{RT}{v-b} n_2 (b_1 - b_2),$$

or

$$\frac{\partial \Omega'}{\partial n_1} = RT \log \frac{v-b}{1+\beta} - \frac{pv}{1+\beta} + \frac{a}{v}\,\frac{1+2\beta}{1+\beta} - 2\beta\, RT\, \frac{b_1 - b_2}{v-b}, \quad . \quad . \quad (d)$$

as $a = a_1$ (see I, p. 769), $\Sigma n_1 = 1 + \beta$ and $n_2 = 2\beta$.

If $\beta = 0$, this passes into

$$\frac{\partial \Omega'}{\partial n_1} = RT \log (v-b) - pv + \frac{a}{v},$$

i.e. the expression (c) for Ω', because then there is only one kind of molecules (viz. complex ones), and hence $\frac{\partial \Omega'}{\partial n_1}$ must be $= \Omega'$, when $\Sigma n_1 = 1 + \beta = 1$.

Now we find for the relation of equilibrium (a) in consequence of (b) and (d), as the function of the temperature C_1 disappears:

$$RT \log \frac{v'-b'}{v-b}\frac{1+\beta}{1+\beta'} - p\left(\frac{v'}{1+\beta'} - \frac{v}{1+\beta}\right) + a\left(\frac{1}{v'}\,\frac{1+2\beta'}{1+\beta'} - \frac{1}{v}\,\frac{1+2\beta}{1+\beta}\right) -$$

$$- 2\, RT\, (b_1 - b_2)\left(\frac{\beta'}{v'-b'} - \frac{\beta}{v-b}\right) - RT \log \left(\frac{1-\beta'}{1+\beta'}\,\frac{1+\beta}{1-\beta}\right) = 0,$$

because $c_1 = \frac{n_1}{\Sigma n_1} = \frac{1-\beta}{1+\beta}$. The accented quantities refer to the *solid* phase. If we now substitute $\frac{v-b}{1+\beta}$ for $\frac{RT}{p+{}^{a}/_{v^2}}$, we find finally after division by RT:

$$\log \frac{p+{}^{a}/_{v^2}}{p+{}^{a}/_{v'^2}} = \frac{p}{RT}\left(\frac{v'}{1+\beta'} - \frac{v}{1+\beta}\right) - \frac{a}{RT}\left(\frac{1}{v'}\,\frac{1+2\beta'}{1+\beta'} - \frac{1}{v}\,\frac{1+2\beta}{1+\beta}\right) + \Bigg\{ \quad (16)$$

$$+ 2\,(b_1 - b_2)\left(\frac{\beta'}{v'-b'} - \frac{\beta}{v-b}\right) + \log \left(\frac{1-\beta'}{1+\beta'}\,\frac{1+\beta}{1-\beta}\right) \Bigg\}.$$

[For the coexistence liquid-vapour, instead of liquid-solid, this relation would pass into

$$log\frac{a/v^2}{p}=\frac{pv'}{(1+\beta')\,RT}+\frac{a/v}{RT}\frac{1+2\beta}{1+\beta}-2\beta\frac{b_1-b_2}{v-b}+log\left(\frac{1-\beta'}{1+\beta'}\frac{1+\beta}{1-\beta}\right)$$

for sufficiently low temperatures, when v (liquid) may be neglected by the side of v' (vapour), $\frac{1}{v'}$ by the side of $\frac{1}{v}$, and $\frac{1}{v'-b'}$ by the side of $\frac{1}{v-b}$, $p+a/v^2$ then being $=a/v^2$, and $p+a/v'^2$ then being $=p$; or it may pass into

$$log\frac{a/v^2}{p}=\frac{a/v}{RT}\frac{1+2\beta}{1+\beta}+\frac{v-(1+\beta)\,b_1}{v-b}+log\left(\frac{1-\beta'}{1+\beta'}\frac{1+\beta}{1-\beta}\right),$$

because $\frac{pv'}{(1+\beta')\,RT}=1$ and $1-2\beta\frac{b_1-b_2}{v-b}=\frac{v-(1-\beta)b_1-2\beta b_2-2\beta(b_1-b_2)}{v-b}=$

$$=\frac{v-(1+\beta)\,b_1}{v-b}.$$

If now moreover $\beta=0$, $\beta'=0$, so that we have to deal with a simple substance, this ($b=b_1$) becomes:

$$log\frac{a/v^2}{p}=\frac{a/v}{RT}+1,$$

VAN DER WAALS' well-known relation for the pressure of coexistence, as viz. a/v^2 may be expressed in p_c and a/v in T_c (see TEYLER, p. 36—37)].

14. Let us return to the coexistence liquid-solid. The formula (16) holding for this might also have been found from the relation:

$$p_0=\frac{1}{v'-v}\int_v^{v'}pdv=\frac{1}{v'-v}\int_v^{v'}\left(\frac{(1+\beta)\,RT}{v-b}-\frac{a}{v^2}\right)dv,$$

in which now the quantity β must be assumed to be variable in the integration between v and v'. But this course would have been far more lengthy, because then we should also have had to make use of the relation of equilibrium (2) [see I, p. 770]. We have, however, convinced ourselves that the result, as might be expected, is identical with (16).

As v and v' may be eliminated by means of the equation of state, β and β' by means of the relation of equilibrium (2), the derived relation (16) is really the required relation $p=f(T)$. But unfortunately these eliminations cannot really be carried out, so that we have to restrict ourselves to deriving the value of the pressure of

coexistence p in the different points of the curve SM (see fig. 6 of the Plate) from the relation (16) in connection with (2) and the equation of state.

With regard to $T=0$, we have already found by another way in II (see p. 35), that then

$$(T=0) \qquad p_0 = \frac{q_0}{-\Delta b} - \frac{a}{vv'} \quad . \quad . \quad . \quad . \quad . \quad (10)$$

For in the relation (2) on p. 26, viz.

$$\frac{\beta^2}{1-\beta^2} = \frac{cT^{\gamma+1}}{p+a/v^2}\, e^{\frac{(p+a/v^2)(-\Delta b)-q_0}{RT}},$$

β will always be $=1$ for $T=0$, when $(p+a/v^2)(-\Delta b) > q_0$ (portion FE of fig. 5 on the plate of III; v is then constantly $=2b_2$). For then the second member $=0\times e^{\infty}=\infty$. If on the other hand $(p+a/v^2)(-\Delta b) < q_0$ (part DC, where v is constantly $=b_1$; and part CB, where v increases from b_1 to ∞), β will always be $=0$ in consequence of $0\times e^{-\infty}=0$.

Along the portion ED of the isotherm, where β changes from 1 to 0 with variable v, $(p+a/v^2)(-\Delta b)-q_0$ must necessarily be $=0$, for else β would have to be either $=1$ or $=0$ according to the above. It is however easy to see, that the mentioned quantity with respect to T must be of the order $RT\,log\,\frac{\lambda}{T^{\gamma+1}}$, for then

$$\frac{\beta^2}{1-\beta^2} = \frac{cT^{\gamma+1}}{p+a/v^2}\,\frac{\lambda}{T^{\gamma+1}} = \frac{c\lambda}{p+a/v^2},$$

which now remains finite for $T=0$, and may yield different values of β for different values of v. In the second member we have $p+a/v^2 = \frac{q_0}{-\Delta b}$ along the mentioned part ED, so that $p+a/v^2$ has a constant value, and for every value of v corresponds a definite value of p. Formula (10) follows then immediately from $p_0 = \frac{1}{v'-v}\int_v^{v'} p\,dv$. And as $\frac{p+a/v^2}{RT} = \frac{1+\beta}{v-b}$, $v-b$ will then be permanently $=0$, hence $v=b=b_1+\beta\,\Delta\,b$, so that β may be found from $\beta = \frac{b_1-v}{-\Delta b}$. (See also the 2nd footnote on p. 120 of III). By comparison of this latter expression for β with the above one the value of λ, left undetermined just now, might be easily found.

15. We now proceed to consider the expression for $\frac{dp}{dT} = \frac{\Delta E}{T\Delta V}$ more closely.

The general equation for ΔV may be found from

$$\Delta V = v - v' = (b - b') + [(v - b) - (v' - b')].$$

In this $b = b_1 + \beta\Delta b$, $b' = b_1 + \beta'\Delta b$, hence $b - b' = (\beta - \beta')\,\Delta b$, so that we get:

$$-\Delta V = (\beta - \beta')(-\Delta b) - RT\left[\frac{1+\beta}{p+{}^{a}/_{v^2}} - \frac{1+\beta'}{p+{}^{a}/_{v'^2}}\right] \quad . \quad (17)$$

Now as $\beta - \beta'$ is always *positive* (we indicated the phase with the *slightest* value of β, i.e. the *solid* phase, by accented quantities), while $-\Delta b$ is supposed to be *positive*, $-\Delta V$ will have its *greatest positive* value at $T = 0$, viz. $-\Delta b$. For then $\beta - \beta'$ has its maximum value $1 - 0$, and the term with RT, in which $\frac{1+\beta}{p+{}^{a}/_{v^2}} > \frac{1+\beta'}{p+{}^{a}/_{v'^2}}$, will be as small as possible. [1])

For ΔE we may write (see (12) on p. 36 of II):

$$\Delta E = (\beta - \beta')(q_0 + \gamma RT) + \left(p + \frac{a}{vv'}\right)\Delta V,$$

or after substitution of the value of ΔV found:

$$\left.\begin{aligned}\Delta E = (\beta - \beta')\,\gamma RT + (\beta - \beta')\left[q_0 - \left(p + \frac{a}{vv'}\right)(-\Delta b)\right] + \\ + RT\left(p + \frac{a}{vv'}\right)\left[\frac{1+\beta}{p+{}^{a}/_{v^2}} - \frac{1+\beta'}{p+{}^{a}/_{v'^2}}\right]\end{aligned}\right\} . \quad (18)$$

Now we saw above that $q_0 - \left(p + \frac{a}{vv'}\right)(-\Delta b)$, i.e. $q_0 - \left(p_0 + \frac{a}{v_0{}^2}\right)(-\Delta b)$, when p_0 is the pressure of coexistence at $T = 0$, and v_0 is the "third" volume on the part $'ED$ (cf. also II, p. 35—36), is of the order

$$-(RT\,log\,\lambda - (\gamma + 1)\,RT\,log\,T),$$

i.e. of the order

$$\alpha T + \beta T\,log\,T,$$

so that the above expression for ΔE must be of the order

$$\alpha' T + \beta\,T\,log\,T = T(\alpha' + \beta\,log\,T)$$

in the neighbourhood of $T = 0$. So it follows from this that for $T = 0$ also $\Delta E = 0$; that for $T > 0$ ΔE becomes at first *negative*

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[1]) For in practice v and v' will differ very little, whereas β will mostly be considerably greater than β' (cf. also II, p. 39 footnote, where we demonstrated on the same ground that $v - b$ is always $> v' - b'$).

(for $\frac{d(\Delta E)}{dT} = (\alpha' + \beta) + \beta \log T$, which has the value $-\infty$ for $T=0$, as β is always positive); then reaches a *maximum* negative value $\left(\text{evidently when } \log T = -\frac{\alpha'+\beta}{\beta}\right)$, increases again for higher values of T, becomes again $=0$ $\left(\text{when } \log T = -\frac{\alpha'}{\beta}\right)$, and then remains always *positive*, and that increasing, because the increase of T and that of $q_0 - \left(p + \frac{a}{vv'}\right)(-\Delta b)$ will exceed the decrease of $\beta - \beta'$ in consequence of the continual decrease of the pressure of coexistence p.

In the expression

$$\frac{dp}{dT} = -\frac{\Delta E}{T(-\Delta V)}$$

$\frac{\Delta E}{T}$ will now be of the order $\alpha' + \beta \log T$ in the neighbourhood of $T=0$, so that then $-\frac{dp}{dT}$ is also of the order $\alpha' + \beta \log T$, because $-\Delta V$ remains finite. In other words: $\frac{dp}{dT}$ is $=+\infty$ for $T=0$: becomes $=0$ for $\log T = -\frac{\alpha'}{\beta}$, where ΔE becomes $=0$ for the second time (see above), and will then become *negative* and continue to *decrease*, because past the minimum of ΔE $\left(\text{for } \log T = -\frac{\alpha'+\beta}{\beta}\right)$ ΔE continues to increase (see above), while we have already seen that $-\Delta V$ is a quantity *decreasing* with T.

So this course of $\frac{dp}{dT}$ gives for the line SM (coexistence liquid-solid) for *negatieve* values of Δb and ΔV a course as represented in Fig. 6 of the plate. (S is the triple-point). Hence the line SMN will *touch* the pressure axis in the point N ($T=0$), because there $\frac{dp}{dT} = \infty$.[1])

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[1]) Disregarding the logarithmical order of ΔE, it has been erroneously derived in II, p. 36 [formule (12a) and (13)] that the limiting value of ΔE for $T=0$ would be γRT, and therefore that of $\frac{dp}{dT}$ would have been given by $-\frac{\gamma R}{-\Delta b}$,

In the point S $\frac{dp}{dT}$ can even become *positive*, viz. when $-\Delta V$ becomes negative. This is e.g. the case when $-\Delta b$ has only a *slight* positive value, so that in (17) the term with $RT\left[\frac{1+\beta}{p+a/v^2}-\frac{1+\beta'}{p+a/v'^2}\right]$ begins to prevail for higher values of T; in other words: when the sign of $v-v'$ is no longer only given by the sign of $b-b'$. In this case the line SM runs as indicated in fig. 7; i.e. with a vertical tangent in A, where $-\Delta V$ changes from positive to negative. Of course this point A may lie at very high pressure, so that it seems that the line SM *continues* to run to the right (which will of course only be the case for Δb positive).

16. It follows from the expression (10) of II p. 35, viz.

$$(T=0) \quad p_0=\frac{q_0}{-\Delta b}-\frac{a}{vv'},$$

which we have already discussed there, that for *small* values of q_0 p_0 (the part ON at $T=0$) may become even 0 and negative. We shall then have a course as indicated in the figures 8—11.

The solid region contracts more and more, when p_0 decreases. At the same time the triple point S will move more and more to the absolute zero point 0.

As soon as p_0 has become negative (fig. 9), there appear necessarily two triple points S and S', as the *realisable* pressure of coexistence remains, of course, positive. So with sufficient lowering of the temperature (the pressure remaining between that of M and S), we get first into the solid region, but finally again into the liquid region.

The possibility of such a course has already been suggested by TAMMANN (see inter alia BAKHUIS ROOZEBOOM, "die heterogenen Gleichgewichte" I p. 83, fig. 9) — with this important difference however, that TAMMANN supposes, besides a vertical tangent in A (see our fig. 7) and a horizontal one in M, another vertical tangent in A' and a horizontal one in M' (see fig. 12). Such a course, however, is

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so a finite negative value; whereas in reality the last limiting value is $=+\infty$. But this error has had no further influence on what follows, as (12a) and (13) have no more been used.

That at first the value of p itself at $T=0$ is *increasing*, appears from

$$q_0-\left(p+\frac{a}{vv'}\right)(-\Delta b)=\alpha T+\beta\, T\, log\, T,$$

which is decreasing for $T=0$, while vv' then remains unchanged.

impossible according to our above theoretical considerations, as for $T=0$ *always* a pressure of coexistence exists (positive or negative). The grounds on which TAMMANN thinks such a course possible, are therefore theoretically not to be justified — as far as the points A' and M' are concerned.

Therefore BAKHUIS ROOZEBOOM has substituted the "half" egg for the "whole" egg of TAMMANN (see loc. cit. p. 93, fig. 10); but such a course (see fig. 13 of the plate) too is incompatible with theory.

BAKHUIS ROOZEBOOM tried to explain with this diagram why in many cases liquids do not *crystallise*, so that with lowering of the temperature a continuous transition would take place from the liquid into the amorphous glassy state, without this latter being metastable.

But this phenomenon is also explained by our theory. For with sufficiently low value of q_0, p_0 becomes smaller and smaller, and even with comparatively very low pressures the solid region (see fig. 9, upper arrow) will no longer be reached. Finally the solid region will quite descend below the line OK and afterwards below the axis $p=0$ (see fig. 10 and 11), and we have a continuous liquid region — viz. from the moment that the line NM touches the line $OS'SMK$ (fig. 10), in which then S' and S will coincide.

Apart from the not accurately indicated course of the line SM past the point M, TAMMANN's figure is therefore closer to the probable truth (indicated by our fig. 9) than BAKHUIS ROOZEBOOM's figure with two pressures of coexistence in B and C at $T=0$. In the latter's diagram (fig. 13) the solid state might sometimes be reached by *increase* of pressure; with us, however (fig. 9), only by *decrease* of pressure.

The *absence* of the solid state can now be ascribed to *four* causes.

1. The liquid mass is already so viscous *before* the melting point (lying on the line SM) is reached, that it passes into the amorphous glassy state. In consequence of this the velocity of crystallisation is so slight, when the melting point is reached, that no crystallisation takes place, at least not immediately. Prof. JAEGER writes me that in such cases sometimes *after a very long time* devitrification takes place. So in this case we can do nothing but wait.

2. The melting point *is* reached in liquid state, but the pressure (e.g. that of one atmosphere), is *too high* (fig. 9, upper arrow). In this case we might try slowly to cool the liquid in a closed glass tube (so that at first it is *quite* filled with the liquid). The liquid is

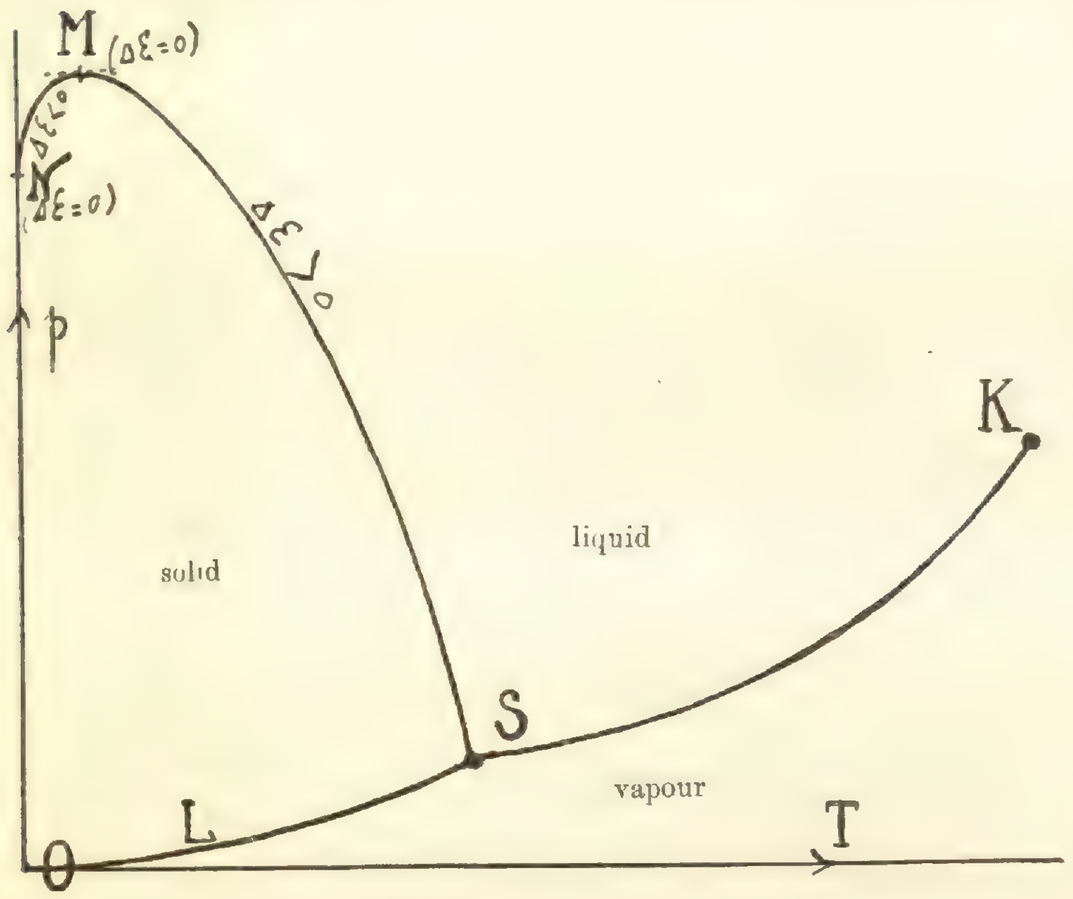

Fig. 6

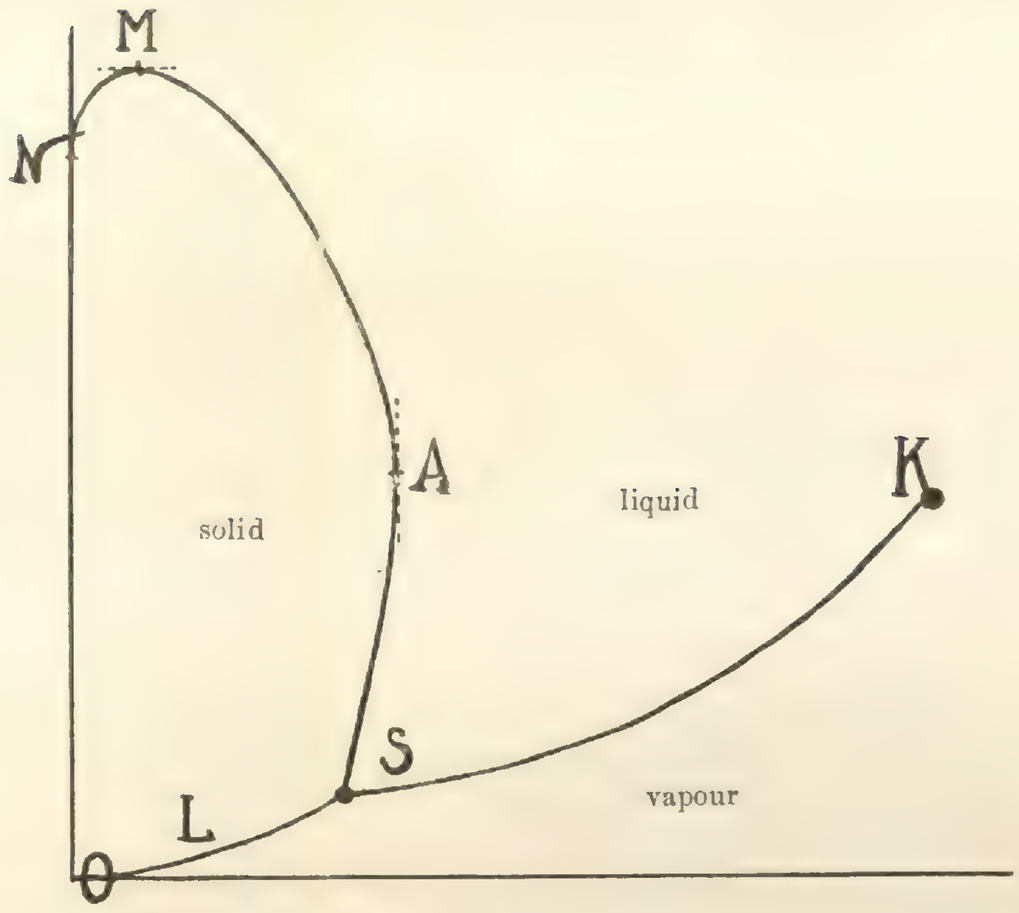

Fig. 7

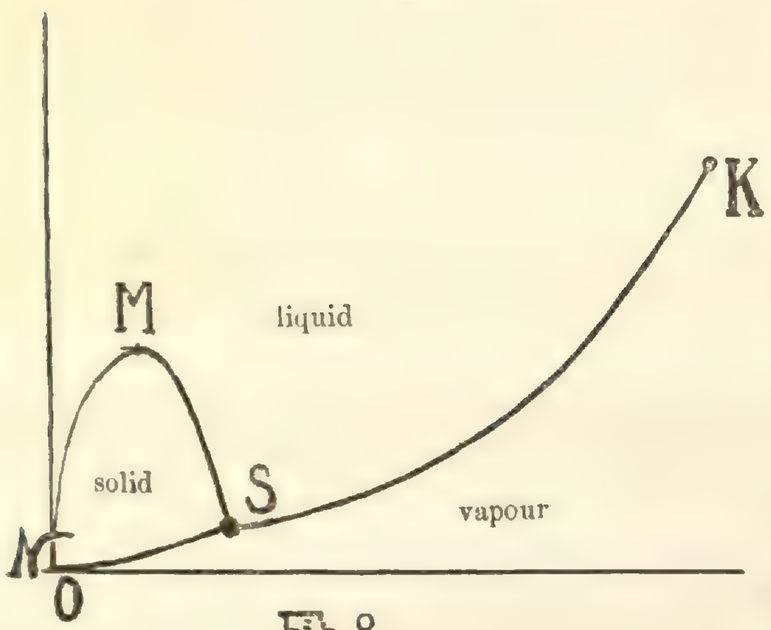

Fig. 8.

$q_0 = \frac{a}{v_0 v_0'}(-\Delta b)\,;\; p_0 = 0\,.$

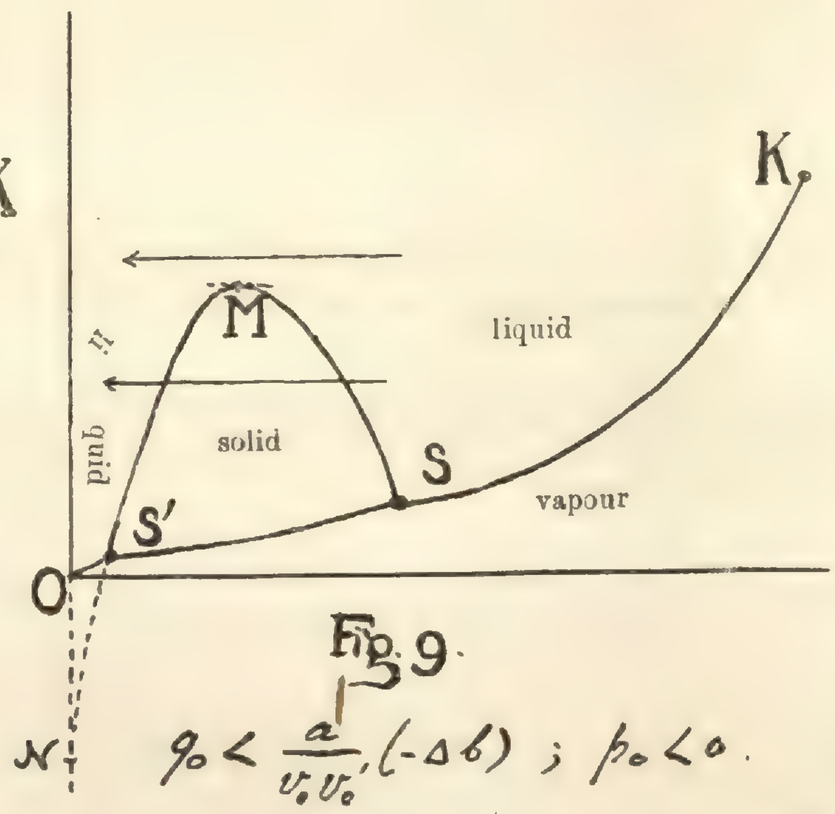

Fig. 9.

$q_0 < \frac{a}{v_0 v_0'}(-\Delta b)\,;\; p_0 < 0\,.$

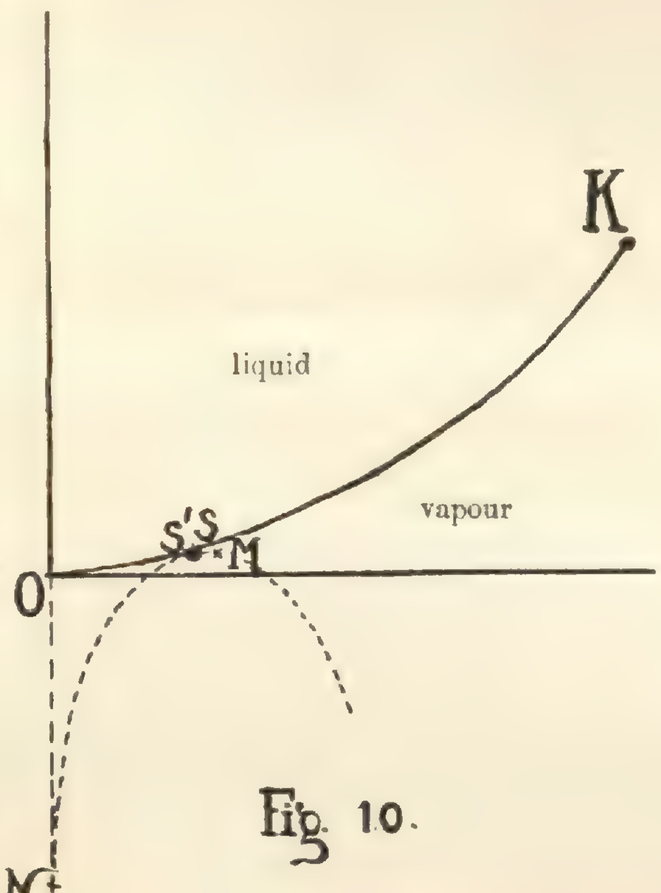

Fig. 10.

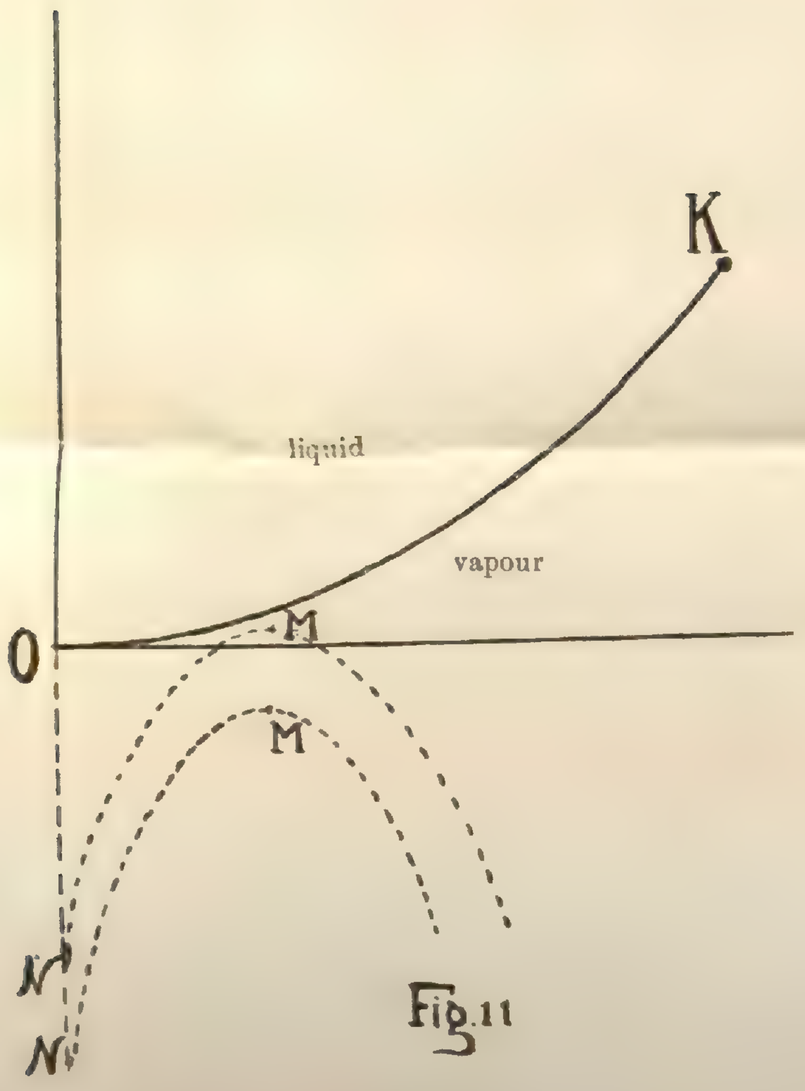

Fig. 11

(TAMMANN.)

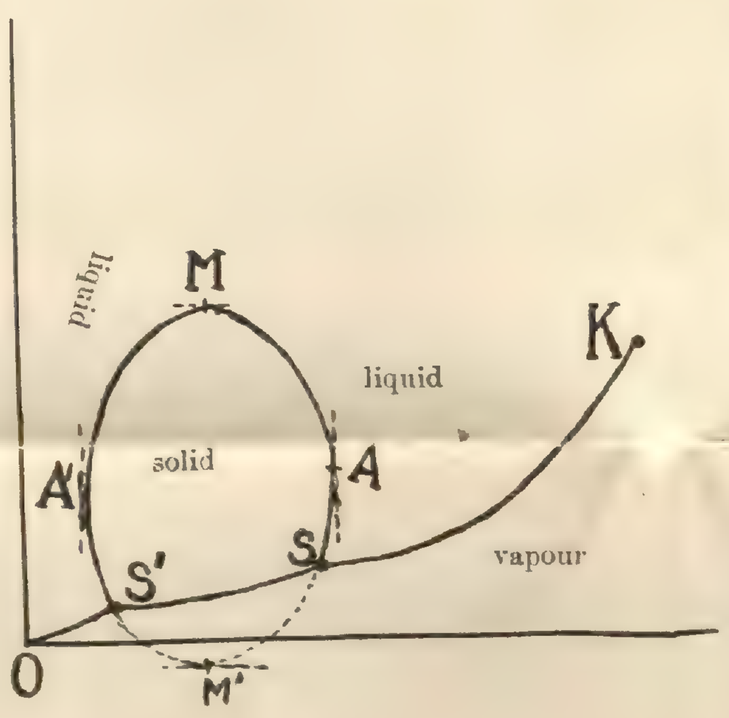

Fig. 12.

(BAKHUIS ROOZEBOOM.)

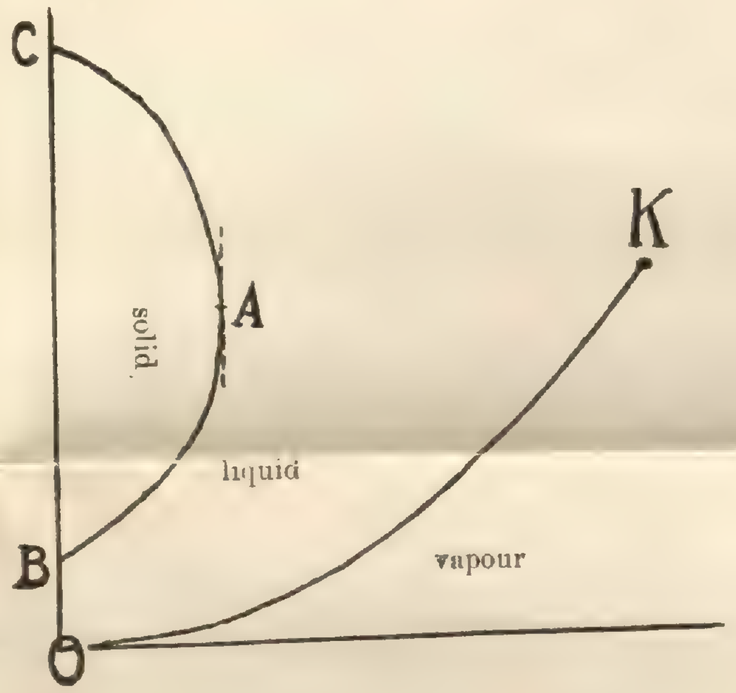

Fig. 13.

then continually under the pressure of the saturated vapour, so that we reach the point S along the line KS. [1])

3. The cooling has not been continued sufficiently long. For in cases as in fig. 9 the triple point S lies mostly at very low temperatures. Then, of course, only the cooling must be continued. It is however to be feared then, that the case 1 occurs before the line SM is reached.

4. There is no crystallized state (fig. 10 and 11). Then we may wait very long before the amorphous glassy mass crystallizes.

(*To be continued*).

Geology. — "*On oceanic deep-sea deposits of Central-Borneo*". By Prof. G. A. F. Molengraaff.

In the year 1894 I discovered in the basin of the Upper Kapoewas in Western Borneo[2]) cherts and hornstones, consisting almost entirely of tests of Radiolaria, which I described as deep-sea deposits. Such rocks are also known as Radiolarite.

The Radiolaria from these deposits were examined by Hinde [3]) and in consequence the age of these rocks was determined as pre-cretaceous, probably as jurassic.

The formation, of which these Radiolarites form part, I named the Danau-formation, after the large danaus or lakes of Western Borneo, where this formation is well-developed and was first observed by me. The Danau-formation there occupies a strip of country of an average breadth of 60—70 K.M. which is bordered on the north side by older formations, namely of the old-slate-formation, while on the south side it disappears under younger tertiary sandstones and volcanic products. The whole formation is strongly folded with an east-west strike, and forms part of the Upper Kapoewas mountain range.

From the lake district I could follow this formation eastward as far as the watershed between the Upper Kapoewas and the Upper

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[1]) Might it not be possible to try to make some of the many substances mentioned to me by Mr. Jaeger crystallise as e. g. several esters of organic acids, amber-acid-nitril, and others?

[2]) G. A. F. Molengraaff. Geological explorations in Central Borneo. p.p. 123 & 414, Leiden 1902.

[3]) G. J. Hinde, ibidem, Appendix I.

Mahakkam rivers and just beyond it, for a distance of 230 K.M. At that time nothing was known regarding the geological structure of the basin of the Upper Mahakkam, but a short time ago I was enabled to resume my studies on the distribution of these Radiolarites since there have been placed at my disposal the collections made by NIEUWENHUIS[1]) in the riverbasins of the Mahakkam and Upper Kajan in the years 1896—97 and 1898—1900, also the unpublished results of investigations made by BONARELLI along the rivers Kèlai and Segah in the Bĕrau district in the years 1905 and 1906, and finally a collection of rocks made by VAN MAARSEVEEN along the river Kèlai and its tributary the Kĕloh in the year 1907. From these studies it results that the Danau-formation extends eastward, without changing its character, from West Borneo into East Borneo right through the basin of the Upper Mahakkam; it reaches the watershed between the Mahakkam and the Kĕlai eastwards of the Boh-river, and is finally also found on the north side of this watershed in the basin of the Kĕlai in the Bĕrau district, always with the same strike from west to east or nearly so. At an average distance of 50 K.M. from the East coast of Borneo it finally disappears in the Bĕrau district under the younger, tertiary coast-range of East Borneo.

The total length of the strip of country, in which the occurrence of the Radiolarian rocks has now been demonstrated, is 650 KM. and if the average width be taken at somewhat over 60 KM. this area occupies roughly 40000 square kilometers. The composition of the cherts and hornstones remains perfectly constant over the entire distance, and in every locality the same types of rocks are found again and again presenting the same macroscopical and microscopical characteristics.

The first type is the Radiolarian hornstone, the true Radiolarite. This rock is semitransparent, hard, brittle and splintery and of a colour varying between milkwhite, red and greenish. It consists for about 97% of silica and is composed almost exclusively of closely packed tests of Radiolaria, joined together by a siliceous cement. The red-coloured varieties may be called Radiolarian jasper; they contain a small percentage of iron. The tests of the Radiolaria are difficult to distinguish under the microscope in the milk-white varieties; they can be much better distinguished in the red jaspery varieties, but best of all they are preserved and discernable in the greenish varieties of the Long Kèloh in the Berau district.

The second type is an argillaceous chert or siliceous clay-shale,

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[1]) A. W. NIEUWENHUIS. Quer durch Borneo. Leiden 1904.

which is always coloured bright red and contains more clay and iron and less silica than the first type. Radiolaria occur in it in variable quantity, but always much less than in the first type.

The two types pass one into the other quite gradually and all sorts of intermediate types occur. The distinct stratification of the cherts of the Danau-formation is caused by thicker strata of pure Radiolarian hornstone alternating with thin layers which contain more clay and less Radiolaria.

Both types and all intermediate stages agree in this respect, that they do not contain any constituents indicative of a terrigenous origin, and that the organic remains they contain all belong to Planeton organisms with siliceous tests, chiefly Radiolaria.

In an uncemented state, in which such deposits are still continually laid down on the bottom of the oceans at the present day, the deposits of the first type are called Radiolarian ooze and those of the second type red clay. Radiolarian ooze and red clay gradually pass one into the other, and only differ in the percentage of organic remains they contain. MURRAY, for instance, places the limit between red clay and Radiolarian ooze at a point where 20% of the deposit consists of tests of Radiolaria. It has been observed, that such deposits only occur in those parts of oceans which are far removed from land, and then again only in the very deepest portions.

This is explained by assuming that at very great depths calcium carbonate is dissolved by sea water, so that of the tests of the Planeton organisms,which continually sink down to the bottom of the sea only the siliceous tests ever reach the bottom in such depths, and consequently they alone are deposited and preserved.

As soon as one assumes that Radiolarian ooze and red clay are oceanic, abyssic sediments, deposited at very great depths, then, on account of the complete agreement, between Radiolarian hornstone and Radiolarian ooze on the one hand, and between red, cherty clay-shale and red deep-sea clay on the other hand, one must also conclude that the cherty rocks of the Danau-formation in Borneo are oceanic, abyssic sediments. It follows then further that at the time when these sediments were deposited, which was probably in the jurassic period, the greater portion of Borneo was the floor of a deep sea far away from land of any importance.

The view, which forms the starting point to this conclusion, and according to which calcium carbonate is dissolved by sea water at great depths, has been subjected to criticism bij THOULET[1]), a criticism

[1]) I. THOULET. Précis d'analyse des fonds sous-marins actuels et anciens p.p. 16 en 33. Paris 1907.

which therefore also applies to the supposition, that Radiolarian ooze is only deposited in the deepest portions of the oceans. THOULET's doubt arose from his observing stratification in some specimens of deep-sea ooze, in such a way that layers of Radiolarian ooze alternated with Globigerina ooze.

PHILIPPI and MURRAY [1]) observed in the specimens of deep-sea deposits collected by the Valdivia, that such a stratification is of common occurrence in deep-sea sediments. They showed that the power to dissolve calcium carbonate belongs especially to the ice-cold water from the polar regions which sinks down at high latitudes and moves very slowly along the bottom of the ocean to the lower latitudes. As this ice-cold water slowly gives off its oxygen it gradually loses its power of oxidizing organic substances into carbon dioxide, and consequently of dissolving calcium carbonate. In consequence the upper limit at which red clay and Radiolarian ooze occur, is deepest at the equator, namely at 5500 M., whereas at higher latitudes in the sub-arctic parts of the oceans, this upper limit is already found between 2000 and 3000 Meters. The position of this limit must therefore be dependent on climatic conditions and in general would be less deep in cold periods with strong development of the polar ice caps, than during warmer periods, when the ice-cold polar water only descends in smaller quantities and at very high altitudes.

PHILIPPI believes that the variations in the extent of the polar ice coverings, which certainly have taken place, explain the stratification of deep-sea sediments in a quite satisfactory manner, and that it is not permissible to doubt the correctness of the hypothesis that at great depths calcium carbonate is dissolved by sea water.

For the present I too find no reason for doubting that Radiolarian ooze and red clay *in equatorial regions* are only formed at great depths, below 5000 Meters, and just as well were formed exclusively at such depths in former ages, and that therefore the Radiolarian cherts and hornstones and the red cherty clay-shales of the Danau-formation in Borneo must have been deposited in an ocean of similar great depth [2]).

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[1]) J. MURRAY and E. PHILLIPPI, Die Grundproben der Deutschen Tiefsee Expedition. Wiss. Ergebnisse der Deutsche Tiefsee Expedition. Bd. X. p. 200, 1908.

E. PHILIPPI. Ueber Schichtbildung am Boden der heutigen und vorweltlichen Meere. Intern. Revue der ges. Hydrobiologie und Hydrographie II. p. 1. Leipzig. 1909.

[2]) E. PHILIPPI. Ueber das Problem der Schichtung und über Schichtbildung am Boden der heutigen Meere. Zeitschr. der deutschen geol. Ges. LX, p. 359, 1908. PHILIPPI suggests that during warm epochs in the earth's history with a mild climate prevailing even in the polar regions, the oceanic sediments laid down on the bottom of the oceans could possess a high percentage of carbonate of lime,

Hitherto not so keen attention has generally been paid to fossil, oceanic, abyssic deposits [1]) as might have been expected, considering that they indicate one of the most extreme situations into which a portion of the surface of the lithosphere can be brought, and considering that 40 % of the bottom of all oceans on earth consist of such deposits, which are still formed there continually.

Probably this is caused by the fact that such sediments have as yet been found in a fossil condition as rocks in but very few places under such conditions as permitting of their character as abyssic sediments being established beyond controversy [2]). This is again intelligible if we bear in mind that only under very special conditions deposits formed at a depth of more than 5000 meters below the surface of the sea, can be lifted so high as to form part of the dry land and become accessible to investigation. Theoretically the only chance of this occurs in mountain chains where sediments, formed at widely different depths, can be forced up by folding to a great height above sea-level.

The conviction that the earth's crust consists of parts of widely different stability, is gaining more and more ground. The more stable portions form extensive blocks which apparently can only change their position relative to each other and to sea-level by very slow upward and downward vertical movements. HAUG [3]) calls these stable blocks, whether situated above or below sea-level, *aires continentales* or simply continents. If they are situated above sea-level the formation of deep-sea deposits is excluded a priori, and if they are situated below the level of the sea, oceanic deep-sea deposits may be formed in case the depth of the sea is sufficiently great, but they have a very small chance indeed to get accessible to investigation.

The more mobile portions of the earth's crust, the geosynclinal areas, separate in broader or narrower strips the stable continental

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even in case they were deposited in abysmal depths. I personally doubt whether climatic changes could bring out such a radical change and consider the explanation given in the next pages for the possible cause of the paucity of fossil, oceanic, abyssic, siliceous sediments more likely.

[1]) Their very existence is even doubted by one of the leading text-books where it is only admitted for Barbados. O. KRÜMMEL. Handbuch der Oceanographie, p. 211. 1907.

[2]) For this a large area of distribution is necessary in the first place, because locally Radiolian rocks can be formed, which are not originated from abyssic sediments. Limestone containing Radiolaria e g. may be locally changed into Radiolarite through silicification and destruction of all organisms with calcareous tests.

[3]) EM. HAUG. Les géosynclinaux et les aires continentales. Bull. de la Soc. Géol. de France. 3. XXXVIII. p. 617. 1900 and Traité de géologie I. p. 157. 1907.

blocks [1]). The movements of the earth's crust which are probably in the first instance a result of the progressive cooling of the lithosphere, are concentrated in these strips. It may be that for a long time they form submarine troughs of very varying depths, the geosynclinals, in which a strong sedimentation and considerable piling up of sediments takes place. Only exceptionally where both the depth is very great and the nearest land surface remote (one can imagine this condition for instance when one or both of the adjoining continental blocks is wholly or partially submerged) oceanic abyssic sediments can be formed in portions of a geosynclinal. Afterwards, however, perhaps in consequence of this considerable sedimentation itself, the conditions of equilibrium of this labile strip may be changed, the geosynclinal may be compressed and thrust up to a mountain chain; the causes of these changes are still very imperfectly known, but in any case the study of the composition of the mountain chains themselves proves that before they were folded up they were geosynclinals. By the folding process the sediments are forced up, at whatsoever depth they might have been formed, and finally they may form part of high mountains [2]). Oceanic, abyssic sediments may in this way come to lie high above the level of the sea, and become accessible to investigation. Thus in mountain chains there is theoretically nearly the only chance, and even that is a small one, that fossil deep-sea sediments come to form part of the dry land. For this reason such deposits will be found limited to mountain chains and be relatively rare on land, however widely they may be distributed at present on the bottom of the oceans, and certainly always have been.

The facts hitherto known completely confirm this theory; all deep-sea rocks, which have so far been discovered occur in mountain chains or at least in geosynclinal areas.

In the first place this applies to the Danau-formation, the largest continuous area of fossil deep-sea deposits which is at present known.

Tectonically the Danau-formation forms part of the Upper Kapoewas mountain range (perhaps it would be better to call this now the Central Borneo mountain range) which extends with an east-westerly strike right across almost the whole of Borneo. The time when this important range was formed, is not yet known with certainty. The

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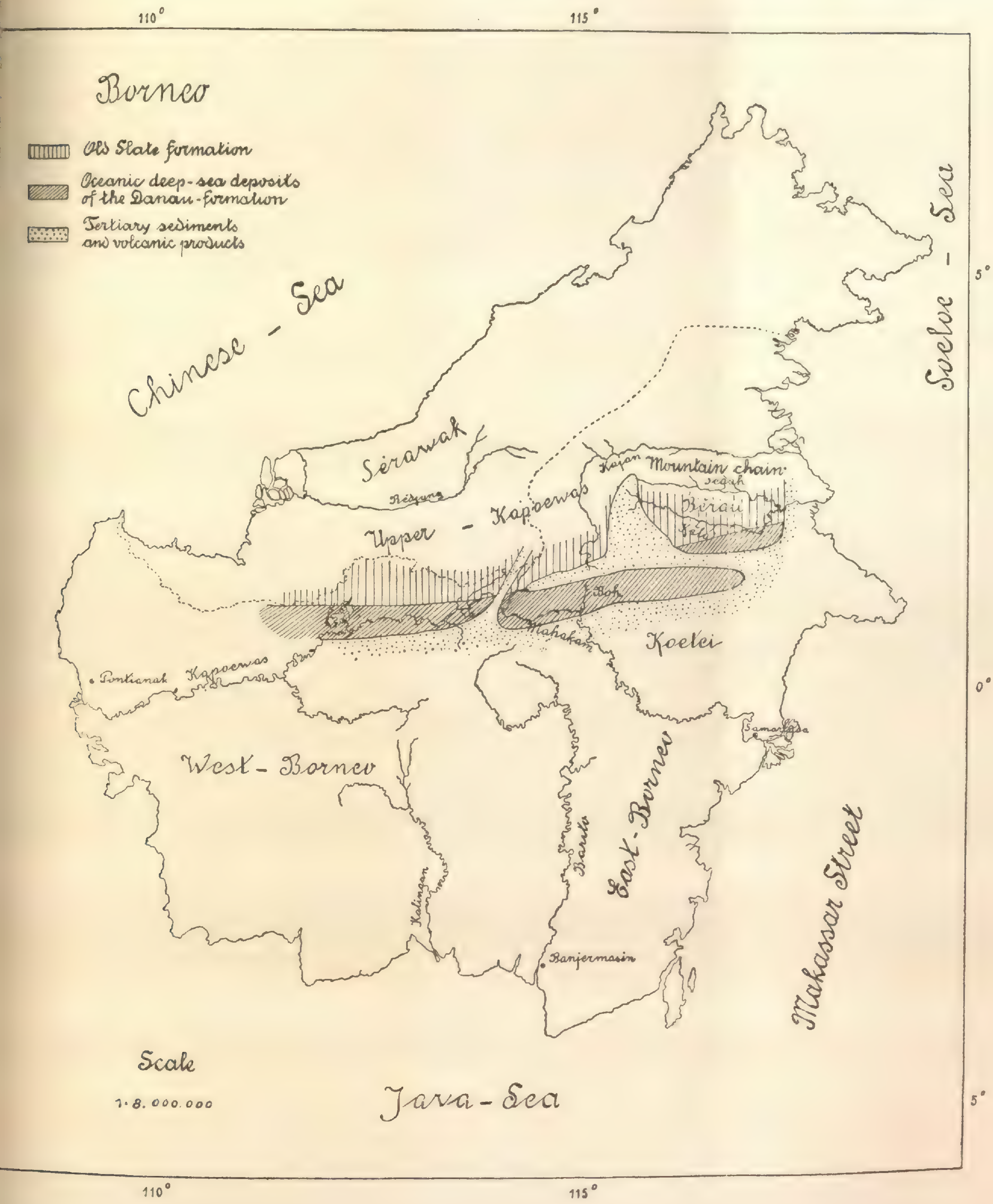
1) The relative position of continents and geosynclinals changes but very slowly and remains permanent during long geological periods.

2) VERBEEK believes that if the folding takes place at great depth, the upper sediments need not be folded too but may be pressed up more or less vertically to a considerable height. In the West Indian as well as in the East Indian Archipelago such movements appear to have taken place on a large scale. R. D. M. VERBEEK, Molukken Verslag p. 816. Batavia 1908.


110°
115°
Borneo
Old Slate formation
Oceanic deep-sea deposits of the Danau-formation
Tertiary sediments and volcanic products
Chinese - Sea
Sulue - Sea
Sérawak
Upper - Kapoewas
Kapoewas Mountain chain
Bérau
Bok
Mahakam
Koetei
Pontianak
Kapoewas
West - Borneo
Kalingan
Barito
East - Borneo
Samarinda
Banjermasin
Makasar Street
Scale
1:8.000.000
Java - Sea
5°
0°

youngest sediments which still have undergone folding belong to the cenomanian epoch whilst the tertiary sandstone which overlies the tilted rocks of the range inconformably is probably of miocene age.

One may assume that the Central-Borneo mountain range probably is the result of the oldest foldings of the Alpine system of crustal movements.

Mesozoic Radiolarian rocks have been described from various other parts of the East Indian Archipelago [1]) e. g. from Ceram, Boeroe, Rotti, Savoe, Celebes, but our knowledge of the tectonic conditions of these islands is still inadequate, and one can only say that the presence of these rocks in our archipelago, which is wholly situated in the area of the so-called Alpine folding system, is not at variance with the theory outlined above.

The upper-jurassic Radiolarian cherts of the Grisons and other parts of the Alps, the jurassic Radiolarian rocks of the Coast Range of California, and also the miocene oceanic deep-sea deposits of Trinidad, Barbados and Cuba, all belong to the same Alpine mountain system, originated during the same period of crustal movements.

The cherts with Radiolaria of the Culm period, which are found for instance in the Harts mountains, in Devonshire, Cornwall etc. form part of a more ancient system of folding, of the Hercynian (variscian-armorican) mountain chain.

The cherts with Radiolaria of the lower silurian of Scotland belong to a still older system of folding i.e. the Caledonian mountain system.

The devonian Radiolarites of New South Wales have been found in palaeozoic rocks probably folded in the carboniferous period.

It is clear from these examples to which various others might be added, that the fossil oceanic, abyssic sediments are exclusively found in strata, which form part of mountain systems due to folding or are in any case limited to the labile portions of the earth's crust, the areas of geosynclinals.

The rarity of these rocks and their peculiar distribution are arguments in favour of the permanence of continents and oceanic basins, but with this restriction that the permanence only applies to such parts of continents and oceanic basins as lie outside the geosynclinals, therefore in favour of the permanence or rather geological longevity of the continental blocks in HAUG's sense.

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[1]) On this the following may be consulted: R. D. M. VERBEEK, Molukken Report p.p. 767, 773, 803 et seq.; and K. MARTIN. "Ein zweiter Beitrag zur Frage nach der Entstehung des Ostindischen Archipels." Geogr. Zeitschr. XIII. p. 425, 1907.

Petrography. — "*On Micaleucite basalt from Eastern Borneo.*" By H. A. BROUWER. (Communicated by Prof. G. A. F. MOLENGRAAFF.)

The occurrence of leucite rocks in our East-Indian Archipelago, at the same time the first occurrence outside Europe, was observed by VOGELSANG from the Gunung Bantal Susum on the island Bawean; VOGELSANG'S observations were described after his death by ZIRKEL [1]) in 1875.

LORIÉ [2]) described leucite rocks from Java from the Gunung Ringgit, an extinct volcano on the north coast of the residence Besuki, and VERBEEK [3]) from the Gunung Muria and the mountain Patti-Ajam in the residency Djapara. BEHRENS [4]) described rocks from the same localities.

In Celebes WICHMANN [5]) discovered in 1888 leucite basalt in the southwestern peninsula among the boulders of the river Pangkadjéné, and leucitites from the rivers Masépé (near the kampong of the same name) and Walannaë, a tributary of the Minralang (upper course of the Tjenrana). RETGERS [6]) mentions leucitites from Malawa (Mountain Regencies, northern district, South Celebes) and finally BÜCKING biotite leucite basalt, underlying limestone with Nummulites east of Kantisang near Bangkang Sakiang [7]), leucite basalt near Tandjing Lossa on the Mandar coast (lat. 2°42′ south) and leucitites in conglomerates near the kampong Tambi in the bay of Manudju [8]).

From Borneo no leucite rocks were hitherto known.

Now Prof. MOLENGRAAFF recently discovered among the collection of rocks, made by Prof. NIEUWENHUIS in the basins of the Mahakam and the Kajan in Eastern Borneo in the years 1896—'97 and 1898—1900, a leucite rock, which he placed at my disposal for further investigation.

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[1]) F. ZIRKEL, Neues Jahrbuch f. Mineral. 1875, blz. 175.

[2]) J. LORIÉ, Bijdrage tot de kennis der Javaansche Eruptiefgesteenten. Dissertation. p. 247. Utrecht, 1879.

[3]) R. D. M. VERBEEK und R. FENNEMA, Neue Geologische Entdeckungen auf Java. Neues Jahrb. f. Mineral. B B. 2. 1883, p. 205.

[4]) H. BEHRENS, Beiträge zur Petrographie des Indischen Archipels. II. Verh. K. Ak. v. W. Amsterkam. XXXIII. 1883.

[5]) A. WICHMANN, Leucitgesteine von der Insel Celebes. Petrogr. Studien über den Indischen Archipel. I. Natuurk. Tijdschr. v. Ned. Indië. LIII. 3. 1893.

[6]) J. RETGERS, Jaarb. v. h. Mijnw. in Ned. Oost-Indië, 1895, p. 99.

[7]) H. BÜCKING, Leucitbasalt aus der Gegend von Pandkadjéné in Süd-Celebes. Ber. Naturf. Ges. Freiburg i. B. 1899, XI, p. 99.

[8]) H. BÜCKING, Beiträge zur Geologie von Celebes. Sammlungen des Geologischen Reichsmuseums in Leiden. VII. 1902—1904, p. 43.

It was found on August 26th 1900 in a boulder bed in the Upper-Kajan above the mouth of the Laja[1]) which is a small rivulet there. The dimensions of the boulder (14 × 8 × 8 cM.) indicate transport over a not very large distance; probably it was derived from the northern slope of the Bawoei Mountains which form the water shed between the basins of the Upper-Mahakam and the Upper-Kajan. A little further to the East, where NIEUWENHUIS crossed this range, it consists of steeply inclined strata of the old slate formation of the Upper-Kapoeas mountains, which are overlaid unconformably by horizontal strata of sandstone.

This leucite rock has the external appearance of a lamprophyr: numerous strongly glittering bronze coloured flakes of mica (±3 mm.) contrast with a very finely crystalline, but not absolutely compact matrix. On careful inspection a very large number of olivine crystals may be seen disseminated through the entire rock; only in exceptional cases do they reach a size of 2 mm.; they are mostly smaller than 0,5 mm. and are noticeable on account of their yellowish brown colour and strong lustre. In external appearance the rock resembles a minette.

Under the microscope the matrix is seen to consist of leucite, large light green microlites of diopside and numerous small idiomorphic crystals of titanic iron ore. The leucite crystals (on the average 0.05 m.m.) frequently show clearly octagonal and hexagonal outlines, others are rounded or have irregular contours; augite microlites included in them are arranged more or less concentrically with their longer axis parallel to the circumference (fig. 4). At a high magnification and by the use of a selenite disc double refraction and twinning laminae can be observed. The diopside-microlites sometimes have somewhat larger dimensions and then appear as elongated light green pleochroitic prisms without well defined terminal faces; they are sometimes twinned. The diopside does not appear as real phenocryst.

The olivine generally forms crystallographically well-defined crystals, with well marked cleavage according to (010); the cleavage according to (001) only indicated by ill-defined short cracks. The olivine has not undergone the slightest alteration, is colourless and transparent and generally free from inclusions; the larger crystals only surround sometimes a few small crystals of iron ore and augite microlites.

The mica occurs in two ways, either strongly resorbed — in which case the ore grains accumulate in a zone just round the crystal, or

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[1]) A. W. NIEUWENHUIS, Quer durch Borneo, II, p. 350. Leiden. 1904.

form a framework within the original crystal of mica arranged in the three directions (fig. 3) — or not resorbed at all.

In the former case hardly any inclusions occur, in the latter case the mica contains very many poïkilitically included leucite crystals and microlites of augite, and thus the mica is in this case the last product of crystallisation. The slightly birefringent sections after the basal pinacoid are not defined crystallographically; they show in convergent light the sharp bisectrix and optical axes diverging with an angle which is unusual for biotite; sections after undetermined directions show twinning according Tschermak's law. The plechroism is strong and abnormal:

c > b > a

pale orange red canary yellow light yellow to colourless.

from which it is evident that also the cleavage laminae are strongly pleochroitic.

The resorbed crystals show, in these parts which have been preserved, the same pleochroism; moreover a continued growth after the resorption period frequently occurs and then the nucleus and the periphery are orientated in the same way (fig. 3).

The isolation of pure mica material was impossible on account of the large amount of inclusions, so that the chemical composition can throw no further light on the abnormal optical properties.

The chemical composition of the rock appears from the following analyses (I) carried out by F. Pisani and the author. For the sake of comparison there are added the analyses of a few other rocks which are closely allied mineralogically and chemically.

I. Mica leucitebasalt. Boulder Oeloe Kajan. Borneo's Ooster-Afdeeling Anal. F. Pisani and H. A. Brouwer.

II. "Minette of Highwood type". Arrow Peak. Highwood Mountains. Montana. Anal. H. W. Foote cf. L. V. Pirsson Bull. N°. 237 U. S. Geol. Survey. 1905, p. 142. (BaO 0.48; SrO 0.25).

III. Missourite. Shonkin Stock. Shonkin Creek. Highwood Mountains. Mont. Anal. E. B. Hurlburt cf. L. V. Pirsson Bull. N°. 237 U. S. Geol. Survey. 1905 p. 115.

IV. Analcime basalt. The Basin. Cripple Creek Distr. Colorado. Anal. W. F. Hillebrand cf. Cross Journ. of Geol. Vol. V 1897 p. 689. (BaO 0.13; SrO 0.12).

V. Leuciteabsarokite. Ishawooa Canyon. Wyoming. Anal. J. E. Whitfield cf. A. Hague. Am. Journ. of Science Vol. XXXVIII July 1889.

VI. Leucitebasalt. Bongsberg near Pelm. Eifel. Anal. E. Hussak cf. E. Hussak Sitz. Ber. K. Akad. der Wiss. Wien Vol. 77 I. 1878.

VII. Biotiteleucitebasalt Pangkadjéné. South Celebes. Anal. Dr. Bruhns cf. H. Bücking. Ber. der Naturf. Ges. zu Freiburg i. B. XI 1899 p. 78.

	I	II	III	IV	V	VI	VII
SiO_2	46.04	46.04	46.06	45.59	47.28	44.35	47.13
TiO_2	2.20	0.64	0.73	1.22	0.88		
Al_2O_3	12.40	12.23	10.01	12.98	11.56	10.20	14.47
Fe_2O_3	3.54	3.86	3.17	4.97	3.52	13.50	13.56
FeO	5.58	4.60	5.61	4.70	5.71		
MnO	sp.	sp.	sp.	0.14	0.13		
MgO	12.60	10.38	14.74	8.36	13 17	12.31	4.16
CaO	8.38	8.97	10.55	11.09	9.20	11.47	9.00
Na_2O	1.62	2.42	1.31	4.53	2.73	3.37	0.81
K_2O	4.87	5.77	5.14	1.04	2.17	4.42	8.00
H_2O	3.55 [1]	2.87	1.44	3.91	2.96		2.94
P_2O_5	—	1.14		0.91	0.59		
SO_3	—	sp.	0.05	0.03			
Cl	tr.	0.11	0.03	0.05	0.18		
Total	100.78	99.76	99.57	99.87	100.08	99.62	100.07

Iddings [2]) already mentions the intermediate place, which the chemical composition of biotite occupies between that of olivine and felspar; the biotite molecule can be split into olivine and the oxide molecules necessary for the formation of felspar or felspatoids. It follows from this, in conjunction with the analyses quoted above, that in rocks of certain chemical composition where the proportions are favourable to the genesis of these minerals, the latter may show a large difference in relative proportions, whilst the conditions of crystallisation are but slightly different. Secondary biotite may be readily formed in these and in closely related rocks.

[1]) Loss on ignition.

[2]) J. P. Iddings. The origin of igneous rocks. Bull. Phil. Soc. Washington. Vol. XII 1892. p. 166.

A leucite basalt, rich in magnesium, is the type from Bongsberg near Pelm in the Eifel (VI), which contains much olivine whereas the forming of biotite shows a certain relation to that of the olivine crystals, for the former are only found deposited round the latter; the complete freshness of both minerals makes an alteration of one into the other improbable; a simultaneous origin is indicated.

On comparison with the "Minette of Highwood type" or "mica basalt" of the Highwood Mountains (II) we see that here the proportion of alkalies to silica by an equal content of $Al_2 O_3$ would have been in itself more favourable for the formation of leucite, whereas this mineral only occurs in a subordinate proportion in the matrix on the side of alkali felspar. This minette contains but little olivine and also has augite among its phenocrysts. The biotite occurs as well in the matrix as also as phenocrysts, and in the latter case the slightly pleochroitic pale yellowish brown crystals are surrounded by a dark border of much stronger pleochroism, corresponding with that of the biotite of the matrix. In this case also there are therefore two distinct periods of mica formation, the first being intratelluric followed by resorption, then resumed growth of the older crystals and crystallisation of the matrix, in this rock however under different conditions, so that the biotite crystals could not attain large dimensions.

With the inverse proportion of alkalies and a high water content the conditions, when rapid cooling under pressure takes place, are favourable for the formation of analcime basalt or monchiquite; the large loss on ignition of the mineral here described makes the presence of analcime in the matrix very probable.

In the biotite leucite basalt (VII), containing but little MgO, which was described by Bücking from South Celebes the biotite occurs as resorbed phenocrysts and here also a second generation of small crystals occurs in the matrix; the crystallographically well defined phenocrysts of olivine have been altered completely into calcite and serpentine. In this case too the leucite is only found in the matrix; the larger crystals (0.1 m.m.) show feeble birefringence and vague twinning laminae. The apatite, which occurs here in fairly large quantities, is entirely absent in our rock. All the other East Indian leucite rocks are poor in biotite (Wichmann only mentions it in the tufas of the Peak of Maros) and in most of them leucite is not limited to the matrix (Djapara).

Chemically the rock here described is distinguished from most other leucite basalts by its lamprophyric character. As in the case of the lamprophyric effusive minerals, separated into a distinct group by Rosenbusch and in which for instance the phlogopite bearing

leucite rocks of the Leucite Hills are classified (Wyomingite etc.), the present rock is characterized by a small Al_2O_3 content, a large proportion of oxydes of bivalent metals, and the preponderance of magnesia over lime, properties which these rocks have in common with the lamprophyric dyke rocks.

Since the origin of mica seems to be connected with pressure and pneumatolysis, so that for this reason already it is rare in the matrix of effusive rocks, it would appear that the occurrence in large crystals as the last product of solidification would exclude a character as effusive rock. That the occurrence of leucite formation is not confined to effusive magmas of definite chemical composition is proved e.g. by its presence in the intrusion of missourite [1]), which is a coarse granular rock consisting of leucite, diopside and olivine with iron ore and apatite. On comparing the analyses I and III we see how much chemical and mineralogical composition are allied; the missourite is the deep seated modification of the rock here described.

Högbom [2]) describes a growing together of leucite and mica from Frascati in which the latter encloses in strongly glittering bronze-coloured crystals several c.m. long, pseudomorphs of leucite which are up to 5 c.m. in length; this is explained by a sudden origin of the large mica individuals, which then enclosed the already crystallized leucites poïkilitically. The complete freshness of the poïkilitical leucite in our rock is peculiar in connection with the instabiiity [3]) of this mineral and the conditions of origin of the mica.

From all that has been said above it follows, that the matrix of the rock here described was very probably crystallized very rapidly and under pressure, conditions which are realized in dykes and small intrusions. Among the lamprophyric dyke rocks our rock is allied to the monchiquites and is distinguished from the minettes not only by the complete absence of felspar but also by the high content of iron ore and the nature of the femic constituents.

At any rate, as long as the geological character of this rock is not determined, it must be ranged into a group with which it agrees in qualitative mineralogical composition; the complete absence of

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[1]) Walther H. Weed and L. V. Pirsson. Missourite, a new leucite rock from the Highwood Monntains, Montana. Amer. Journ. of Science 1896, II, p. 315.

[2]) A. G. Högbom. Uber einige Mineralverwachsungen. 8. Verwachsung von Leucit und Glimmer. Bull. Geol. Institution of the University of Upsala. Vol. III, p. 450. 1896—1897.

[3]) A. Lacroix. Etude minéralogique des produits silicatés de l'éruption du Vésuve (Avril 1906). Paris 1907. p. 96.

felspar as well as the abundance of mica and olivine is sufficiently expressed in the name micaleucite basalt.

It is left to further explorations of this interesting territory, to make out to what extent the above mentioned suggestions are correct, and whether this rock belongs to a group of lamprophyric dyke rocks, of which the transitions to minettes and monchiquites are known in their leucite bearing varieties.

EXPLANATION OF FIGURES.

Fig. 1. Above, a phenocryst of olivine; below a resorbed crystal of mica and to the left of this non-resorbed mica in the position of greatest absorption with well-defined cleavage lines. The matrix consists of leucite, augite microlites and ore.

Fig. 2. From the top somewhat to the left to the bottom somewhat to the right the dark band consists of non-resorbed mica; the poïkilitical character with regard to leucite and diopside microlites is clear. The olivine encloses a few ore granu'es and augite microlites.

Fig. 3. Resorbed mica, surrounded by a border of similarly orientated non resorbed mica (continued growth of the crystals after the resorption period).

Fig. 4. Leucite crystals with inclusions, poïkilitically surrounded by non-resorbed mica.

Chemistry. — *"On the radioactivity of Rubidium compounds."* By Dr. E. H. Büchner. (Communicated by Prof. A. F. Holleman).

Although the experiments to be described are far from completed, I think it all the same desirable to make a provisional communication as to the results already obtained, in view of the long time these kinds of experiments require.

Campbell [1]) discovered about two years ago that potassium salts, although but very feebly so in comparison with uranium and the other radioactive salts, emit rays which ionise the air and so cause the discharge of an electroscope. This discovery was confirmed from various quarters and Levin and Ruer [2]) succeeded in observing the action of potassium salts on the photographic plate. It seems, therefore, to be pretty certain that potassium salts are either radioactive themselves, or are always contaminated with a radioactive substance; but as it has been proved that this impurity cannot be one of the known active substances it is natural to assume the existence of an active alkali metal, which should always exist in traces in potassium salts. In order to gradually solve this question, it seemed, in the

1) Le Radium IV, 199.
2) Phys. Zeitschr. 9, 248.

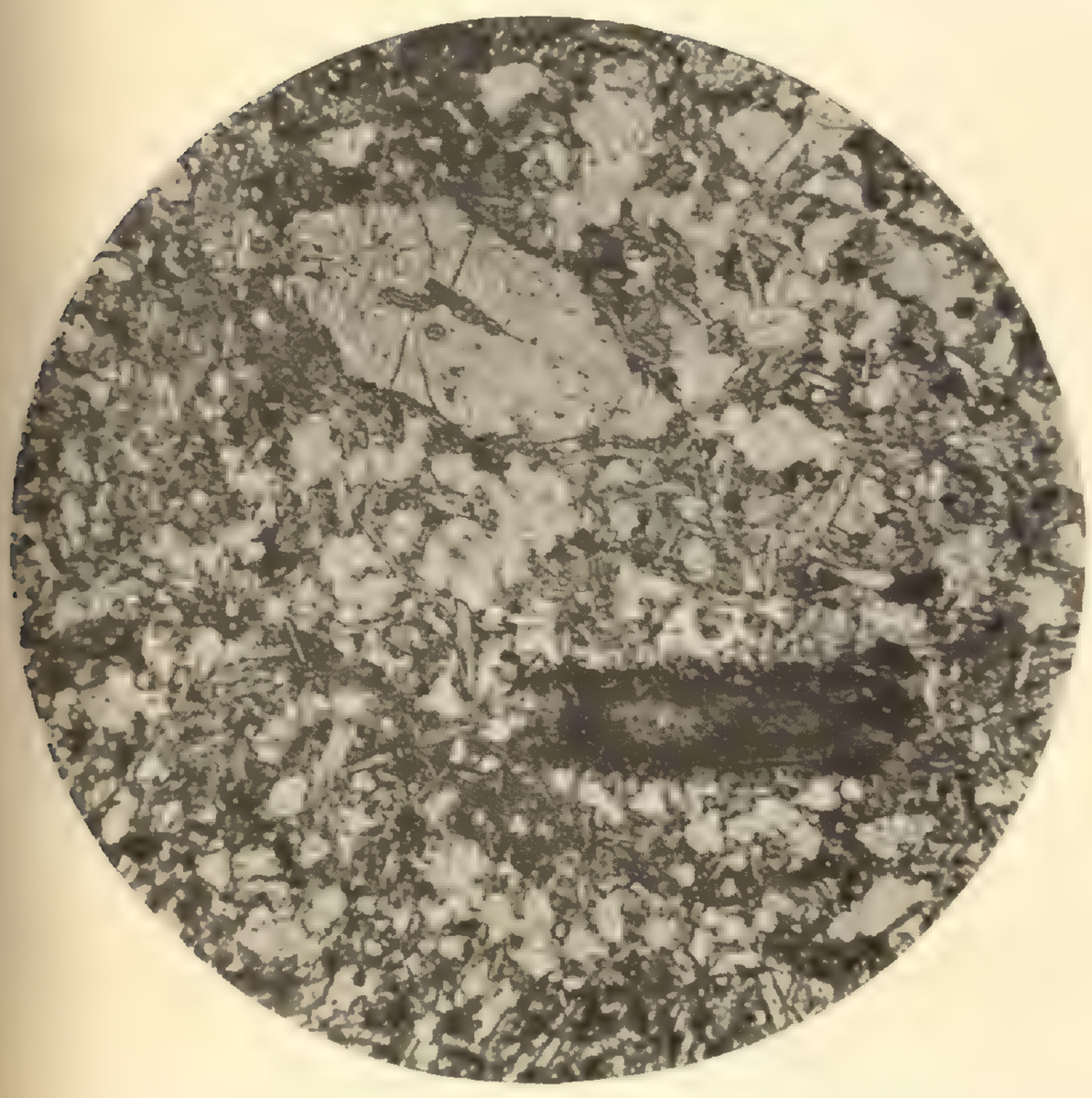

Fig. 1.

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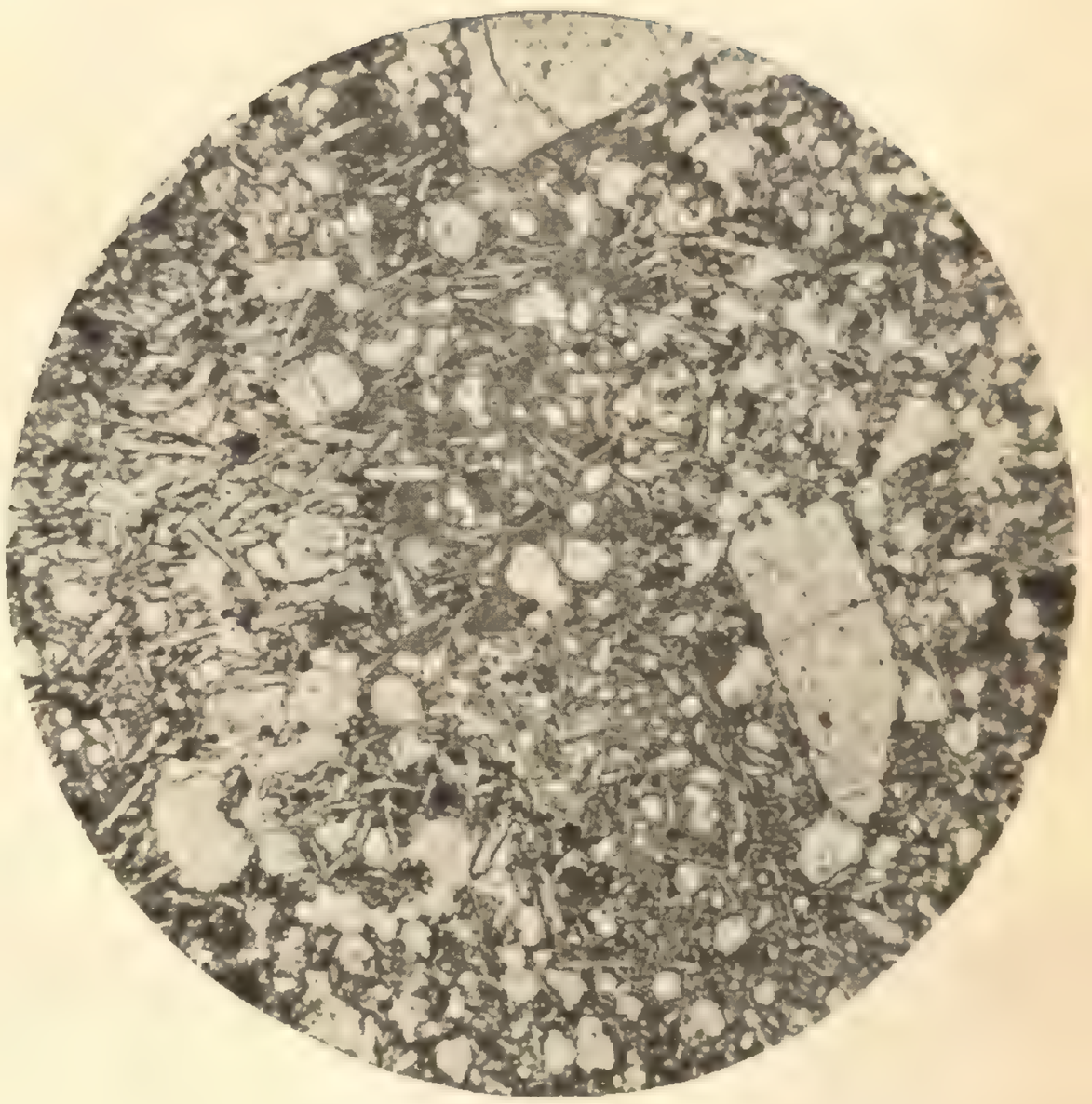

Fig. 2.

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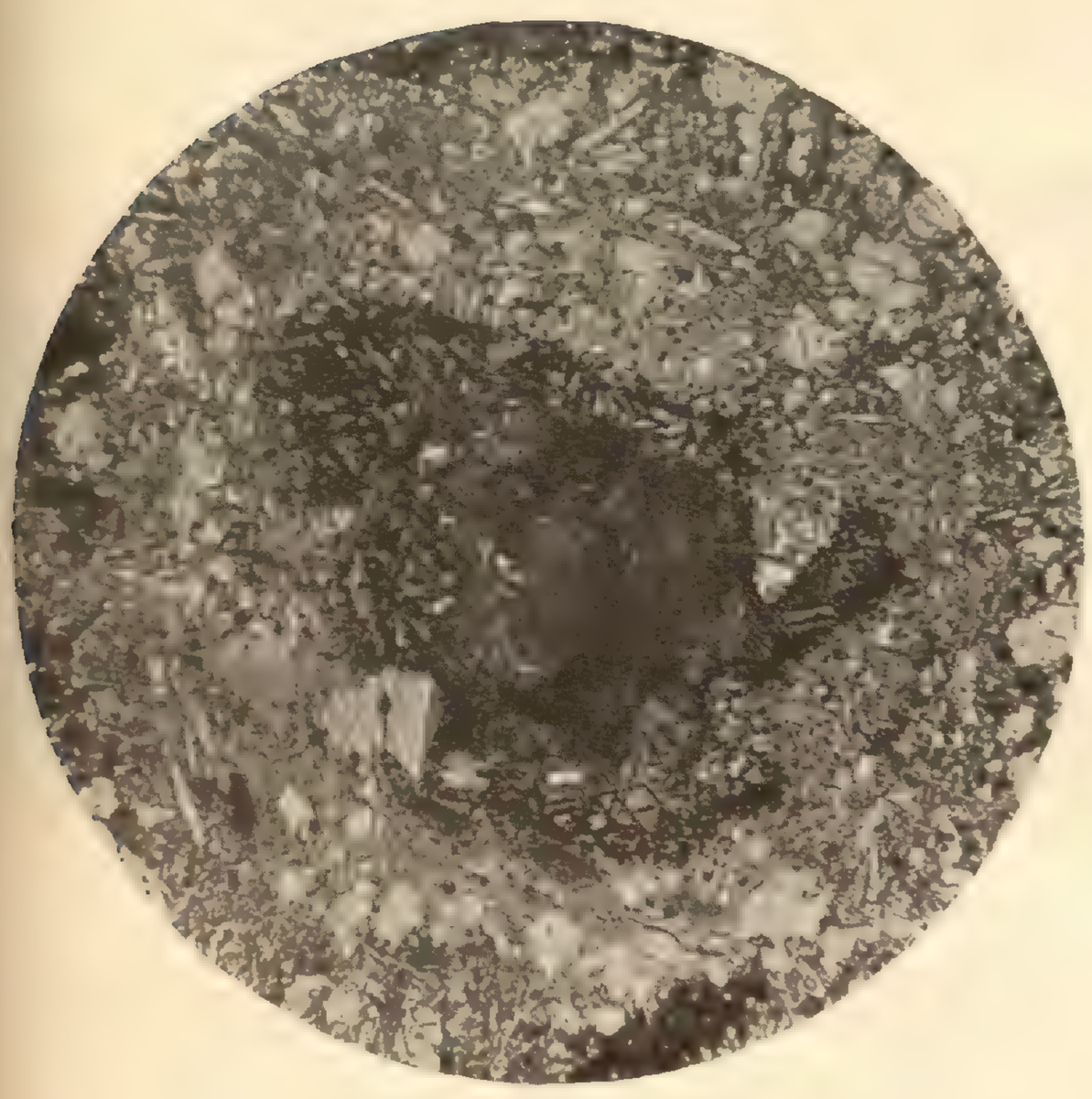

Fig. 3.

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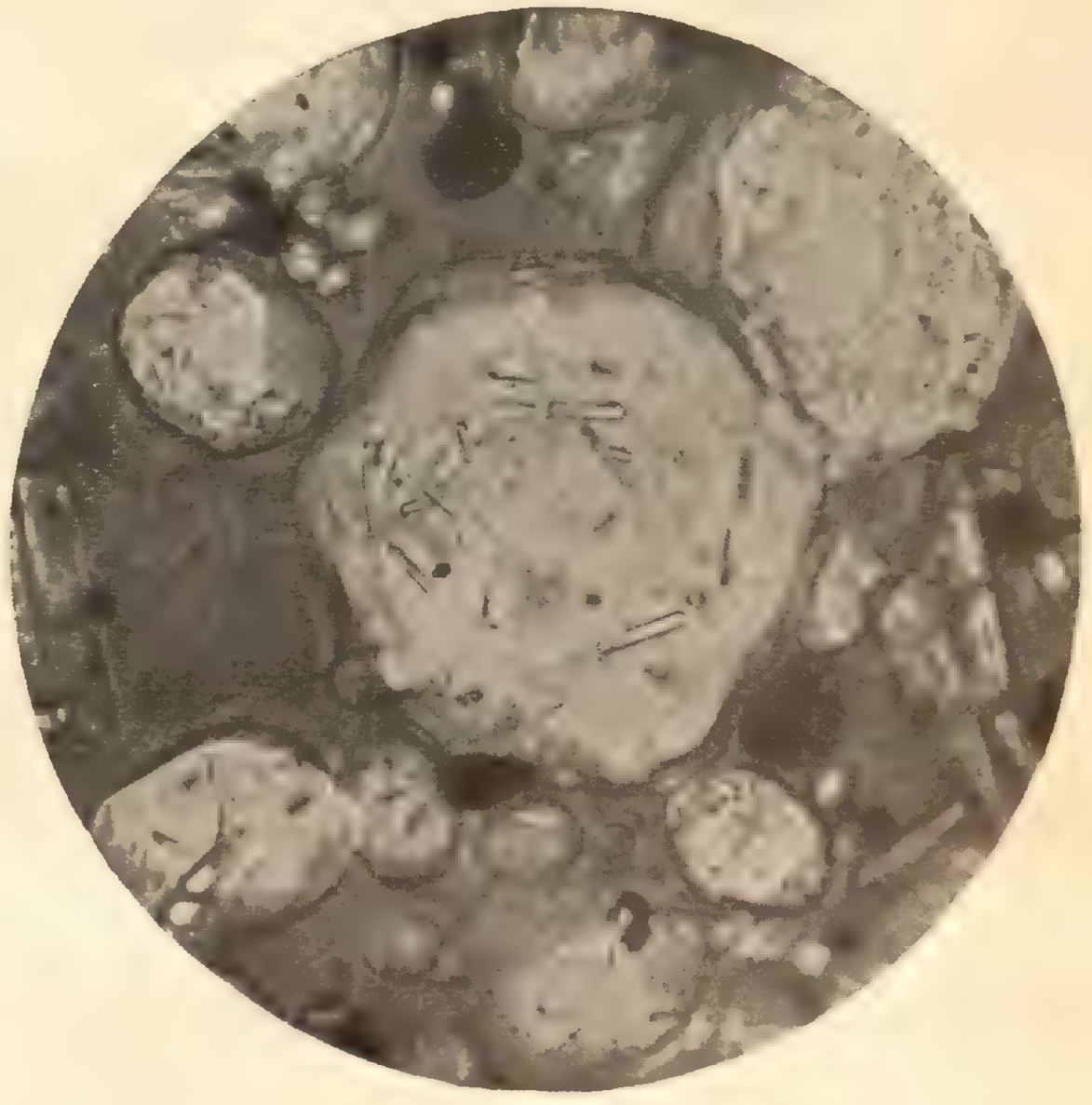

Fig. 4.

500 ×

first place worth while to ascertain, whether other alkali salts have the same property as the potassium compounds. Particular attention had to be paid to the salts of rubidium and caesium, as these are isomorphous with potassium salts, and if there should really exist an unknown alkali metal, it was to be expected that this would also separate together with Rb- and Cs-salts.

Experiments in this direction have been made, partly before and partly during my investigation, by CAMPBELL [1]) and MCLENNAN [2]). By means of the electroscopic method they proved that rubidium had an activity of the same order as potassium, whereas compounds of Cs, Li, and Na did not appear to be active. This last result was confirmed by my first experiment; from a series of photographic plates which were exposed to the action of K_2SO_4, Rb_2SO_4, Cs_2SO_4, NaCl, and Li_2CO_3 the only one which was found to be altered after development was that exposed to the rubidium.

In the following experiments, I have provisionally left out the Cs and Li and have only worked with Rubidium salts, and for the sake of comparison, with the potassium and sodium salts. The experiments were conducted as follows: LUMIÈRE-plates were wrapped up in black paper into which was cut a circular hole in such a manner that the opening came on the sensitive side of the plate. The plates, emulsion downwards were then put on strips of cork, between which was placed an object glass with the salt to be examined. As the salt was just under the opening in the black paper an eventual radiation from the salt had to penetrate only 3—4 mM. of air before reaching the photographic plate. Three plates at a time were now placed in a large box into which was also put a dish with calcium chloride, and the whole after being wrapped up in black paper was placed under a bell-jar over calcium chloride and kept in a dark room. The various salts were used in the anhydrous state and on opening the boxes they appeared to be quite unaltered. All the plates were developed and fixed in exactly the same manner. The result of the experiments was that after exposures of 81 and 117 days, the plates exposed to K_2SO_4 and Na_2SO_4 were quite unaltered whereas a black spot was noticed on the plate exposed to the Rb_2SO_4. The phenomenon although weak is not in the least doubtful.

Another series of experiments were then made in a somewhat different manner; the plates were again wrapped up in black paper with an opening, but this was now covered with aluminium foil

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[1]) Proc. Cambr. Phil. Soc. 15, 11 (from an abstract).

[2]) Le Radium V, 142.

0.001 mm. thick. On this aluminium was now placed very carefully à quantity of the various salts so that the action must proceed through the aluminium. After the end of the experiment the foil was carefully examined for any holes formed but none were noticed; for the rest the arrangemen was the same as before. After an exposure of 97 days the same result was obtained as in the former experiments; the rubidium sulphate had caused a black spot on the plate but the other salts had not affected the same. The effect of the rubidium sulphate was, however, weaker than in the previous experiments.

The general result, therefore, is that rubidium exhibits a stronger action than potassium or sodium, both of which had during the time of the experiment no marked influence on the plates. (In one experiment with potassium plates there probably was an action but so faint that it must be considered doubtful). As regards sodium, this agrees with the results of previous investigators; as regards potassium, however, it does not. This difference may, perhaps, be attributed to the fact that they made use of rubidium-alum, which only contains about 16 % of rubidium whereas I worked with the anhydrous sulphate which contains about 64 % of the metal. But it is also possible that the difference is caused by a different method of working, for it is known that when comparing radioactive substances quite different results are obtained when one works first with the electroscope and then with the photographic plate. In view of this, experiments according to the electroscopic method are in course of preparation. Finally, a third possibility should be considered. It might be possible that Rb_2SO_4 only then acts on the plate when it has been exposed previously to the light, in which case there would be no question of radioactivity, but the remarkable result of the experiments would consist in the fact that the action should be due to the rubidium salt only. In order to come to some decision the following experiment was made. Rb_2SO_4 was dissolved in water, in the dark some drops were placed on a glass plate and after evaporation to dryness the plate was heated for some time at 150°, this leaving a residue of completely dry Rb_2SO_4. This was now covered with aluminium foil and a photographic plate was placed on it at a distance of 0.5 m.m. After 100 days, the plate was developed just like a plate which had been placed in the box in exactly the same manner but without being exposed to the salt. The same result was again obtained; a black spot was visible only on the plate exposed to the rubidium.

This result seems to show that rubidium emits indeed a radiation

which does not have its origin in an energy previously absorbed from outside sources. I intend, however, to repeat this experiment with $Rb_2 SO_4$ which has been kept in the dark for three months, and am also thinking of experimenting with other rubidium salts. Until then I will refrain from drawing further conclusions.

Physics. — "*The behaviour of the phosphorescent sulfides of the alkaline earths at various temperatures, and particularly at very low temperatures.*" By Prof. P. LENARD, Prof. H. KAMERLINGH ONNES and Dr. W. E. PAULI. Communication N°. 111 from the Physical Laboratory at Leiden.

§ 1. For two reasons it seemed to us of importance that the behaviour of the "phosphors" [1]) of the alkaline earths should be investigated at the lowest possible temperatures. In the first place, former experiments of LENARD and KLATT [2]) in which the temperature was lowered to — 180° C., had already shown that the development of many emission bands in these "phosphors" would end only when the temperature was lowered still further, and it was an open question if that further development should correspond with the views already deduced ("1904", pp. 666 *et seq.*) concerning the change with temperature in the properties of the phosphorescence bands. Temperatures so low as those which may be reached with liquid hydrogen seemed to offer particular facilities for clearing up this point, for in other phenomena peculiarities which can be attributed to the fixing of electrons become prominent just at these temperatures [3]).

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[1]) For the sake of brevity we shall in this paper refer to a phosphorescent substance as a "phosphor".

[2]) P. LENARD and V. KLATT. Ann. d. Phys. 15, p. 451 *et seq.* 1904.

This paper which contains very numerous experimental data, and which has already indicated the main points of the theory here given concerning the processes involved in phosphorescence, will henceforth, for the sake of brevity, be referred to as "1904".

[3]) The changes in the increase of electrical conductivity of metals with lowering of temperature noticeable at very low temperatures, in correspondence with the idea first expressed by KELVIN, that the conductivity attains a maximum value at very low temperatures (cf. KAMERLINGH ONNES and CLAY: Comm. N°. 95^c, 95^d These Proc. Sept. '06, 99, Sept. '07, 107^c, Nov. '08), were regarded by KAMERLINGH ONNES (Comm. Physic. Labor. Leiden Suppl. N°. 9 p. 27 1904) following the views of P. LENARD, as a consequence of certain places (dynamids) in the atoms having the power at very low temperatures of holding electrons which, at higher temperatures, are in free motion. The different conditions of motion of the electrons between and around the dynamids were visualised as three states of aggregation

In the second place, the suppositions concerning the mechanism of phosphorescence which were developed in "1904" could be put to the test and extended, if we succeeded in observing any influence of the magnetic field upon the phosphorescence bands. That such an influence cannot be detected at ordinary temperature has been known for years by P. LENARD, who for this purpose investigated Ca Bi-, Ca Mn- and Ca Ni-"phosphors" in strong magnetic fields. And as all bands at ordinary temperature are very broad and ill-defined there seemed to be no chance of making further discoveries in this direction. Now, however, it had appeared from the above mentioned experiments in liquid air, that all phosphorescence bands become remarkably narrower and sharper as the temperature is lowered ("1904" p. 455), and it seemed to be worth while examining if perhaps, at the temperature of liquid — or otherwise of solid — hydrogen the narrowing of the bands should continue so far, that a further investigation in the magnetic field would afford a new outlook upon the problem.

Indeed, the researches of H. and J. BECQUEREL and H. KAMERLINGH ONNES upon the phosphorescent uranyl salts, completed last year and published [1]) after the present investigation had been commenced, were successful in this respect.

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and it was remarked that the magnetic field should exert a similar influence as that exerted by temperature upon the transition of the electrons from one state to another.

This image was further developed in later researches by J. BECQUEREL and H. KAMERLINGH ONNES (Comm. Nº. 103, Febr. 1908, particularly pp. 595, 597—598, 602) on the absorption-spectra of compounds of the rare earths and their behaviour in a magnetic field, which were undertaken also on account of the particular influence of liquid hydrogen temperatures upon the effect of forces exerted by ponderable matter upon the electrons. As "gas or vapour" state can be distinguished that state of motion in which the electrons can occasion conduction of electricity through the atoms when these are in metallic connection, by "liquid", a state of motion in which they are sensitive to light-vibrations and absorption phenomena, while, on the other hand, a "solid" state is a state of motion in which they can take part neither in conduction, nor in absorption of light.

Interpreting the results of the experiments just mentioned in terms of this theory we find that by the circumstance that at very low temperatures the sphere of influence of the dynamids extends to a greater distance the free path in the electron vapour is greatly diminished, and that hydrogen temperatures are already sufficiently low to cause the electrons at various parts of the atom to assume the solid state; these are just the places where the electrons at temperatures that are not so low are particularly numerous in the liquid state, as can be seen from the occurrence of various absorption bands.

[1]) H. and J. BECQUEREL and H. KAMERLINGH ONNES, Comm. Physic. Lab. Leiden. Nº. 110. These Proc. May 1909.

We have therefore investigated a series of well-defined "phosphors" of the alkaline earths as described in "1904" in liquid and solid hydrogen, — hence to a temperature of 14° K. The general result we first communicate here: The bands behaved quite as we expected in becoming narrower and sharper, so that the wave-lengths of their maxima could be determined at these low temperatures under very favourable circumstances. But they remained bands whose intensity did not support a strong dispersion, and this is the reason that we have for the present postponed the continuation of the experiments in the magnetic field. Our aim has rather been to study the particular temperature properties of a number of definite bands, and from these we have attempted to draw general conclusions concerning the process of phosphorescence. In this we have found the ideas already develóped fully supported; in certain respects, however, some not unimportant extensions of these conceptions have been obtained and hence we publish our results here in full.

§ 2. *Apparatus.* The apparatus shown in fig. 1 was used for the experiments with liquid hydrogen. The liquid hydrogen is introduced [1]) through the german-silver tube a to the transparent vacuum glass b, which is inserted in a vacuum glass c containing liquid air. The gaseous hydrogen is led away through d to the gasometer or the vacuum-pump; e is a safety tube, and at the same time a manometer. The german-silver tube f attached to the german-silver covering by means of a packing tube allows a vertical or a rotational motion to be communicated to the rod $g_1 g_2$.

By means of θ_1, a steel capillary which enters the opening of the hollow rod and is there soldered, the rod $g_1 g_2$ carries two copper plates p_{11} and p_{21} which are provided with small frames p_{12} p_{13} on the longer sides. In the copper frames p_{12} thin glass plates q can be fitted, and on these the powdered "phosphors" are scattered in a layer of varnish [2]) and fastened under a very low pressure. The front of the frame is provided with numerous incisions so that the glass plates can be clamped by bending down the little teeth formed in the process of cutting, without exposing the "phosphors" to pressure. The two plates p_1, p_2 and the screw and nut at the upper end of the smooth rod g enable one to observe different

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[1]) Cf. Comm. N°. 103, Pl. I. fig. 4; Comm. N°. 94*f*, Pl. III, Sept. '06.

[2]) We satisfied ourselves that the varnish used did not phosphoresce even at —260° C. At temperatures that were not so very low we had thought of using lanoline for attaching the "phosphors", but it seemed to give rise to an afterglow which in one of our substances almost led us upon a false track.

"phosphors" under the same conditions and immediately after each other.

Between the two plates p_1 , p_2 lies the reservoir θ_0 of the helium thermometer which indicates the temperature of the "phosphors": it is an elliptical cylinder of german-silver. It is attached to the steel capillary θ_1 which, enclosed in g_1, passes through the tube f, and by this means is connected with the glass capillary θ_3 which allows readings for accurate determinations of temperature between — 260° C. and — 180° C. and with the reservoir θ_4, on which temperatures higher than —180° C. can be read by approximation. The purpose served by the valves θ_5 and θ_6 needs no further explanation.

To the bottom of the hydrogen glass b is brought a german-silver warming coil s_0, whose leads are insulated and made airtight in the cap h. This coil serves to vaporise the hydrogen; when this has vaporised the temperature of the space b slowly rises, and by this means the simultaneous change of the phosphorescence and of the temperature can be followed.

In order to work at the temperature of solid hydrogen, the hydrogen gas is pumped away by the vacuum-pump along d, and the pressure is lowered until a crystal layer is formed on the liquid hydrogen; after the crystal layer has grown to a certain thickness it is kept in that state by regulating the rate at which the gas is pumped away.

As the source of light for exciting phosphorescence an iron-arc was used with or without the Wood ultra-violet filter (arranged as in "1904" p. 244) which had been very suitable in former experiments, and also a carbon arc with the same filter and concentrating lenses. Ultra-red light which often proved serviceable, was obtained from the same arc lamp by using as light filter a layer of water and a layer of a solution of iodine in carbon bisulphide. We found no noticeable absorption of this light in the layers of liquid air and liquid hydrogen which it had to traverse. In order to put this to the proof we so manipulated the height of the surface of the liquid hydrogen that the rays falling upon the "phosphor" passed only through glass and gas, and the "phosphor" came just above the surface of the liquid and no more, so that although in the vapour space, it remained just as strongly cooled; in these circumstances we observed not the slightest change.

The substances embraced by our experiments were the "phosphors" Ca Cu Li, Ca Mn Na (yellowshade), Ca Ni Fl, Ca Bi Na, Sr Zn Fl, Sr Mn Na, Sr Bi Na, Ba Cu Li, Ba Bi K. They were all prepared so

that they showed those bands, which will be discussed in the sequel, well developed. Phosphorescent zinc sulphide, uranyl fluoride fluorammonium, and a crystal of uranyl nitrate were also examined.

§ 3. *Temperature phases.* An ample and comprehensive treatment of the change of light emission of the "phosphors" with temperature has already been given [1]).

According to that exposition of the subject, to each band of the phosphorescent light there belong three phases, which as the temperature falls succeed each other in the following order: 1. the upper momentary phase, or heat phase, 2. the permanent phase, 3. the lower momentary phase or cold phase. For each of the various phosphorescence bands the range of temperature embraced by the permanent phase, which also determines the two other phases, lies at a definite height upon the scale of temperatures. In the lower momentary phase the electrons which under the photoelectric influence of the illumination are ejected from the metallic atom are fixed and stored [2]) in the neighbourhood [3]); in the upper momentary phase this fixing of the electrons does not occur for they return immediately causing emission of light; in the intervening permanent phase some of the electrons return and others are fixed in the neighbourhood. It is this permanent phase in which the "phosphor" shows the long and bright afterglow of its respective band. In the earlier experiments which reached a temperature of only — 180 C. the existence of the three phases remained hypothetical for a number of bands, but we are now in a position to show that for these bands the apparently missing regions of permanency and of the lower momentary phase can really be attained by a further

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[1]) "1904"; also P. LENARD: Verh. des Naturh. Med. Ver. Heidelberg, 5 Febr. 1909, pp. 9, 10 *et seq.* We shall in the sequel refer to this comprehensive paper as "1909".

[2]) All the ejected electrons are not thus stored: some of them return at once and in this way cause the momentary light during the illumination; see in this connection § 7 of this communication. This "momentary process" happens with more or less intensity (according to the exciting wave-length) alongside the second process of storing the electrons. It was in a former paper ("1904" p. 667) considered as an extreme possible case that finally in the lower momentary phase the momentary process should take place alone without any storing of electrons. In no single band, however, has this been since realised, and therefore on the contrary the storing of electrons has been chosen in "1909" as the characteristic of the lower momentary phase.

[3]) By this expression we always mean parts of *the centrum itself*; it has already been shown in "1904" (p. 671) that the material which surrounds the centra has nothing to do with the retaining of the charge; cf. also "1909", pp. 14 *et seq.*, and § 7 of this communication.

lowering of the temperature. Thus while up to the present we observed only the momentary bands Ca Mn β, Sr Mn β, Ba Cu β_3, we have now observed the permanent phase and the lower momentary phase, and in Ca Ni β, Ca Bi γ and Ca Bi α_2, SrZnα, Ba Bi α_2 the hitherto unobserved lower momentary phase. Thus the prophecy formerly made ("1904" p. 483 note 2) concerning these "phosphors" has now been found to be fulfilled [1]).

As a general rule one can say about the temperature phases at very low temperatures, that *the temperature of solid hydrogen* (in round numbers —260° C.) *is low enough to bring all bands of the "phosphors" of the alkaline earths* [2]) *into the lower momentary phase.* As the temperature falls from higher values to that of liquid air (—180° C.) new bands continually appear in the permanent phase. Also at the temperature of liquid air various bands are of long duration, the most striking being the persistent and clear afterglow of the CaNi-"phosphors" (CaNiβ band) ("1904" p. 435). At all lower temperatures, however, down to —260° C. one can observe but a small continuous afterglow; only the orange-yellow afterglow of the BaCu-"phosphors" is here conspicuous for its persistence and clearness, having a maximum at —190° C. All other bands either appear in the lower momentary phase in so far as they have not already gone beyond it, or they pass through a region of permanency lying very low and of small intensity (such as BaCu β at —240° C., CaMn β at —250° C.). At the still lower temperature of —260° C. all phosphorescence becomes exclusively momentary [3]).

In all the "phosphors" investigated exposure to cold has resulted in discovery of only one new band; it is a green band in CaBi lying not far from the green heat band CaBi β, but contrary to this it is to be indicated as the lowest cold-band which assumes a state of permanency at —255° C, although, as has already been remarked in general concerning these temperatures, the intensity is very small.

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[1]) The permanent yellow-red band of the SrMn-"phosphors" which had formerly remained unnoticed on account of the smallness of the range of the permanent phase was now found to be at —60° C.

[2]) In the present communication we have left out of account the "phosphors" of lead, silver, antimony and also of SrCu.

[3]) A thermoscope that would serve many purposes both at low temperatures and at others up to red-heat, could be made out of a number of small pieces of different "phosphors" enclosed in a glass tube devoid of air. These should be so chosen that the range of permanency of their bands at different points of the desired temperature interval should end as sharply as possible. The temperature could then be determined according to whether they glow or not with a definite colour when exposed to light.

If we compare the "phosphors" of the alkaline earths with respect to their temperature phases with the uranyl salts investigated by H. and J. BECQUEREL and H. KAMERLINGH ONNES, we find that in the latter case no results were observed which would oppose the theory that the emissions of the uranyl salts also undergo three temperature phases. Only, the permanent phase and, *a fortiori*, the lower momentary phase must then lie at temperatures much lower than the temperatures reached by those experimenters and by us. To their results we may add that uranyl fluoride fluorammonium behaves just the same as the other uranyl salts examined by them, while in the case of uranyl nitrate we have confirmed their results. The emissions of the uranyl salts have thus been observed only in the upper momentary phase, in which no storing of the electrons takes place; were one able to bring them to the lower momentary phase (below — 260° C.), then, after illumination at that temperature, one would be able to observe a glow as the temperature rises, a result attempted but not attained by the above-mentioned experimenters at the lowest temperature reached at present [1]).

§ 4. *On the glowing of the "phosphors" as the substance is warmed from low temperatures.*

It was proved "1904" p. 458 that as a "phosphor" is warmed after excitation by light those bands appear in succession, the temperature ranges of whose permanent phase succeed each other on the scale of temperatures. This appears to be the case also in the range of temperatures here investigated. If we carefully compare the glow occasioned when passing through various temperatures with the afterglow at various constant temperatures, we find that a "phosphor" that has been exposed to illumination responds as long as the temperature is within the limits of the range of the permanent phase of the band in question. That affords us a means that has proved of great service to us, of determining the range of the permanent phase for bands for which it had not previously been known. For this purpose the "phosphor" was illuminated at the temperature of solid hydrogen, and was then slowly warmed by means of the warming coil *s* shown in Fig. 1, and in the meantime it was watched until the glowing band for which the temperature range of the permanent phase was examined, lighted up. The temperature interval

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[1]) According to § 7 of this Communication, in the case of the emissions of the "phosphors" of the alkaline earths it also depends upon the wave length of the light used to excite the phosphorescence whether we obtain storing and therefore responsive glow, or not.

within whose limits this occurs is the region of permanency of the band investigated. In this manner various experimental results given in this communication were obtained.

Thus the region of afterglow of the band BaCu α_2 extends from $-$ 260° C. to $+$ 350° C. [1]), and is therefore unusually extensive; the region of warming-glow of this band is equally wide. An example of a narrow range on the other hand is given for the yellow band CaNi β from $-$ 180° C. to $-$ 100° C., and in this case the warming-glow is restricted to within the same narrow limits.

The storing of the effects of illumination is at the temperature of solid hydrogen particularly large and unusually complete. If, after illumination of the phosphorescent substance at $-$ 260° C. it is slowly warmed, it remains nevertheless quite or nearly quite dark; but when the lower limit of the permanent phase is passed, the emission of light begins almost at once with great intensity. It is just the non-appearance of the effects of the illumination in the lower momentary phase that makes it comprehensible that the storing should reach such a great value. Its value remains, however, beneath a certain limit. If the illumination in liquid hydrogen is continued longer and longer, the intensity of the warming-glow finally ceases to increase. This must be explained by the circumstance that as soon as the centra which occasion the bands have obtained their full charge they adopt other periods of oscillation (corresponding with the periods of the light which can extinguish the phosphorescence [2])) and could not thus in any case absorb any more of the exciting light.

With regard to the intensity of the light during the illumination in the lower momentary phase, we found that for a definite exciting wave-length, *e.g.* the light passed through our ultra-violet filter, the various bands behaved in widely differing manners. Bands that at 260° C. are of very small intensity in the lower momentary phase are *e. g.* Ca Mn α, Sr Mn α, while others such as Ba Cu α_2, Ba Bi α_2, radiate with great clearness under the same circumstances. The latter, however, also lose their light quickly after illumination — a behaviour corresponding with the known properties of the lower momentary phase — and show also the very perfect storing which we have just mentioned. Later (§ 7 of this Communication) we shall bring these different behaviours of the various bands under one general point of view, by making the fundamental assumption that the quantitative relation between the momentary process and the simultaneous continuous

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[1]) At temperatures above the ordinary, the intensity of both the responsive glow and the afterglow with ultra-red excitation is indeed very small.

[2]) "1909". Also P. Lenard and Sem Saeland Ann. d. Phys. 28 p. 499, 1909.

process in one and the same band differs according to the wave-length which gives rise to the processes.

§ 5. *Response to illumination by long waves* (*red and ultra-red*) *and subsequent extinction.* It has already been shown ("1909", p. 7 [1])) that the action of ultra-red upon the "phosphors" consists of a local molecular rise in temperature of the centra. If the temperature of the centrum rises so far that it enters the permanent phase region of temperature, then the substance begins to respond — previous excitation being understood — just as if the whole "phosphor" had been brought to the higher temperature. When the stored charge is used up, darkness follows the glow, and it is extinguished. It has also been already shown that when ultra-red light is the acting agent a different local molecular rise in temperature belongs to each band, also to those of the same "phosphor", and that each active metal has a particular distribution of extinguishing powers over the spectrum. Discussion of these characteristics in themselves will be reserved for a special communication; our main wish for the present has been to concentrate our attention upon the confirmation and extension of these views which have been developed from the results of observations obtained at very low temperatures.

Corresponding with the great charge which is kept stored at low temperatures, the response to ultra-red at these temperatures is also very intense. As a general rule in this case the actual *lighting up* is far more striking than the subsequent extinction which here takes place far more slowly than at ordinary temperature. We may just now instance the extreme case of zinc sulphide, in which the lighting up which always precedes extinction ("1909") escaped the notice of even a careful observer [2]). If one allows ultra-red light to act upon zinc sulphide in liquid hydrogen, one observes a glow during many seconds [3]). As other examples of response to ultra-red in liquid hydrogen we may name: Ba Bi K, extremely intense and remaining green with the α_2 and β band, hence like the afterglow at ordinary temperature, Ba Cu Li very clear, red, α_1 band, hence like the glow at ordinary temperature; and similarly Sr Bi Na with clear bluish-green responsive glow. To

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[1]) Also P. LENARD and SEM SAELAND *loc. cit.* note 2. p. 164.

[2]) A. DAHMS, Ann. d. Phys. 1903.

[3]) The colour is first green, and then reddish. By projecting a spectrum on the "phosphor" immersed in solidifying hydrogen we have found that the red responsive-glow lies more in the proximate part, the green more in the ultra-red of the spectrum.

ultra-red in liquid hydrogen those bands respond as a general rule the range of whose permanent phase does not lie too high. So in the case of Sr Mn Na the yellow-red band β_1, together with a little of the green band α. The region of the permanent phase for the band β_1 lies about —60° C., or very approximately between —100° C. and —45° C., that for the band α, between —100° C. and +100° C. Hence the local temperature of the β_1 centra had not in any case risen far above —45° C., nor that of the α centra far above —100° C. The same can be seen even in liquid air. In a mixture of carbon dioxide and ether (about —50° C.) on the other hand the "phosphor" responds with a clear yellow light (α band) just like the afterglow at ordinary temperature, and then becomes dark; in this case therefore the local temperature of the α centra has already risen above +100° C. From this it can be seen that the local temperature attainable with a given intensity of ultra-red depends upon the temperature of the material filling the space surrounding the centra. Apparently the centra attain that temperature at which the energy communicated to them by the ultra-red is equal to the energy given in the same time to the surrounding material in the form of molecular motion; an equilibrium is established just as in the case of warming any body which in the same times loses an equal quantity of heat.

In this way it has already been explained in "1909" p. 18 that different kinds of centra in the same "phosphor" undergo at the same time different increases in temperature, according to the greater or less efficiency of their insulation from communicating energy to the surrounding material. Except for these internal characteristics, the local temperature depends upon the intensity of the ultra-red rays, as we saw just now was the case with the temperature of the surrounding material *i. e.* of the "phosphor". The following will serve as an example of the dependence of the phenomena upon the two latter circumstances. Ca Bi-"phosphors" prepared specially for this purpose, show next the blue α group, whose permanent phase ranges not far above + 200° C., also a green β band whose permanent phase lies in the neighbourhood of + 300° C. The insulation of the energies of both kinds of centra seems to be practically the same, for when the ultra-red rays are strong enough, it responds first blue, + 200° C., and then green, + 300° C. If the intensity of the ultra-red rays is now small enough only the blue is seen to respond, and apparently the local temperature of + 300° C. can now be no longer reached. Now increasing the intensity of the exciting rays causes the green to appear. When

the same experiment was carried out in liquid hydrogen, we were unable even with the most intense ultra-red to get the green to appear; in this case only the blue appeared, and hence the local temperature of + 300° C. cannot be reached when the temperature of the substance is in the neighbourhood of —250° C.

§ 6. *Spectral distribution of excitation and temperature.* In a former communication ("1904" p. 471) the supposition was advanced that the excitation-distributions corresponding with the various phosphorescence-bands were independent of the temperature. At that time, however, only the region extending from ordinary to higher temperatures had been investigated. Now, however, in a mixture of carbon dioxide and ether (about —50° C.), in liquid air (—180° C.) and in some cases also in liquid hydrogen (—250° C.) an intense spectrum has been thrown upon the "phosphor" spread out flat, and by this method of illumination the result given above was confirmed. Sr Cu Na, Sr Zn Fl, Ca Mn Na, Ba Cu Li were investigated, the last "phosphor" in liquid hydrogen. Here it appeared that the characteristic excitation-distribution which is shown in Pl. III N°. 39 *loc. cit.*, and which reaches far into the visible region, holds good at that just as at ordinary temperature. In this case the spectrum (from a carbon arc) exciting the phosphorescence had to pass through two vacuum glasses (cf. fig. 1), and by this some of the finer details were necessarily destroyed. In the mixture of solid carbon dioxide and ether and in liquid air, the "phosphors" were enclosed in flat quartz tubes and placed about 1 mm. below the free surface of the bath; the spectrum of a quartz amalgam lamp was then projected upon it from above through a quartz prism; in this way there was less opportunity for disturbance. Here, then, there remains only the great difficulty to which attention has already been drawn ("1904" p. 464) that the exciting spectrum is discontinuous, so that in a certain measure only the rougher features of the charge distribution always appear, while further finer characteristics must necessarily be obscured by the line-grouping in the spectrum which is used to excite the phosphorescence. It is these rougher features of the excitation-distribution — which were already reproduced Pl. III "1904" [1]) — that remain unaltered also at low temperatures.

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[1]) One of these excitation-distributions, viz.: that of Ba Cu α_2 (Pl. III N°. 40) must — as has since been discovered — undergo a correction. We intend later to deal in a separate paper with a new investigation of the excitation-distribution by means of a continuous spectrum.

§ 7. *Momentary and permanent parts of the spectral excitation.* There had still remained unsolved a question concerning the excitation-distribution in the spectrum which aroused our great interest, and in this respect a new result was obtained.

A characteristic behaviour in the distribution of the exciting power in the case of the clear green head band α of the Sr Cu-"phosphors" had already been noticed ("1904" p. 470; shown graphically by the curves 1 and 2 in fig. 2 and reproduced from Pl. III N°. 21 *loc. cit.*); the distribution of the excitation over the spectrum did not disappear regularly, it separated into two spectra, the one quickly (momentarily) increasing and decreasing, the other of much longer duration overlapping this, consisting of completely different groups, and slowly increasing and decreasing. By observing the gradual disappearance, and also the similar increase in intensity, one immediately gains the impression that here two different processes proceeding with different velocities overlap, each of which, however, has a different excitation spectrum ("1904", p. 470).

Then (although mentioning the other view that is now confirmed) the band was regarded as consisting of two coincident bands or of two bands lying very close to each other, $\alpha_1 + \alpha_2$, one of which at ordinary temperature should be in the momentary phase, and the other in the permanent phase, and which, as is always the case with different bands, should each have its own excitation-distribution over the spectrum. The same characteristic of the increasing and decreasing of the excitation-distribution occurring not as a whole as is the case with the Sr Cu α band, has later been mentioned by A. WERNER [1]) in the case of the Sr Zn α band, and a more detailed investigation of a great number of other bands repeated by P. LENARD with better apparatus [2]) than those employed before showed that the above mentioned property is characteristic of all bands, and that it differs in the various bands only quantitatively. As well as these two bands, Ca Mn α_1, Ba Cu α, Ca Bi β are also striking examples of this. The great number of instances observed makes it improbable that in every case one is dealing with the overlapping of bands that lie very close together and are in different temperature phases.

The following observations for which different temperatures were chosen lead to the same conclusion. The observations at low temperatures were made according to the method already described; for those

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[1]) A. WERNER. Dissert. Kiel. July 1907. Ann. Phys. 24 p. 190, 1907.

[2]) Which were rendered available by the establishment of the Radiological Institute at Heidelberg.

at higher temperatures and up to the beginning of red heat, the "phosphor" enclosed in a flat quartz tube was placed in a hot-air bath and illuminated by a spectrum. From these experiments it appeared that those parts of the spectrum to whose excitation momentary response was given and which we shall call collectively the "momentary part", *remained* momentary as far as low temperatures up to those of liquid air [1]) were concerned at *all* temperatures, while those portions of the spectrum which induced an afterglow, the "permanent part", induced this afterglow only within the temperature range of the permanent phase for the band in question, as was to be expected. At the temperatures of the upper or of the momentary phases also the "permanent part" becomes momentary in accordance with the properties of this phase.

It is especially worth noting that whenever the bands are in the permanent phase, the particular momentary part overlapping the "permanent part" nevertheless remains unchanged, so that when a band is excited even at temperatures within the permanent phase, the two processes which we have called "momentary" and "permanent" proceed simultaneously [2]) in such a way that the quantitative relation between the two processes differs according to the exciting wave-length. Some wave-lengths induce almost the momentary process alone, others almost only the permanent process; in general, any wave-length will induce both processes at the same time. For instance, $\lambda = 400^{\mu\mu}$ excites Sr Cu α momentarily, $\lambda = 434^{\mu\mu}$ permanently; $\lambda = 200^{\mu\mu}$ and $\lambda = 450^{\mu\mu}$ excite Sr Zn α momentarily, $\lambda = 366^{\mu\mu}$ permanently; in the same way $\lambda = 200^{\mu\mu}$ excites CaMnα momentarily and $\lambda = 334^{\mu\mu}$ permanently. The general characteristic of the excitation-distribution is that the shortest wave-length (in the extreme ultraviolet) induces only the momentary process, and that towards the limit of the excitation-distribution on the side of the greater wave-lengths continuous and momentary excitation interchange periodically one or more times [3]).

If the permanent part in the afterglow is to be well observed, illumination must be continued for several minutes with a very intense spectrum, for it has always appeared from the experiments that a powerful ("1904" p. 477) and also a prolonged ("1904"

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[1]) In liquid hydrogen the observation as already indicated § 6 proved too difficult.

[2]) This point has already been elucidated particularly with reference to the Sr Cu α band ("1904" p. 668). By special measurements A. Werner has demonstrated it for the Sr Zn α band.

[3]) A separate communication will deal with these characteristics.

p. 281) illumination is necessary to excite permanent emission to the full. If the illumination has not been sufficiently prolonged, or has been performed with a spectrum that was not sufficiently intense, it can happen that places of moderate duration will at first show a clearer afterglow than places of maximum duration because the latter had not yet been sufficiently excited.

As has already been said, the momentary part always seemed to overlap the permanent part, sought for in this way. The former can be observed during the first illumination with the spectrum, or better still, in that part of the phosphorescent light which quickly disappears immediately after the illumination ceases. The momentary glow that was to be observed in these parts of the momentary portion, *e.g.* in the extreme violet, showed in all cases the character of the *upper momentary* phase, that is to say, there was no storing of electrons; parts illuminated by this portion of the spectrum cannot be made to glow either by warming or by ultra-red excitation, only those parts of the spectrum which belong to the permanent part will induce a glow.

There are thus definite wave-lengths which in all temperature phases of the centra bring into action the momentary process exclusively, or almost so; that is to say, the electrons that under the photoelectric influence of these wave-lengths are ejected from the metallic atoms of the centra return quickly to their metallic atoms, giving out light, while at the same time other wave-lengths cause electrons to be ejected from the metallic atoms of the same kind of centra, and these electrons are retained in the neighbourhood (storing, process of afterglowing). *This circumstance can be ascribed to the fact that the ejection from the metallic atoms caused by the various wave-lengths takes place in different directions in space (fixed in the atom), and therefore also in different directions with regard to the neighbouring atoms of the centrum,* the atoms of the alkaline earth metal, the sulphur atoms, perhaps also atoms coming from the additional substance.

Only those electrons which are directed towards places in the neighbourhood suitable for retaining electrons, are retained and stored, thus causing the permanent phase as the temperature falls within the region of permanency. It has already been indicated where these places must be looked for whose presence makes storing possible. It has already been shown in "1904" that they can only be places *within* that molecule to which the name "centrum" has been given. "For every band (kind of centrum) even of the same "phosphor" stores ("1904" p. 473 *et seq.* p. 481) up its own excitation in

such a manner that it cannot be used by any other band" [1]), and also the storing of electrons (the permanent phase) in the different bands of the same "phosphor" is often bound to widely differing temperatures ("1904" p. 761); this has also been further confirmed by our experiments at the lowest temperatures. The material surrounding the centra, which is the same for all kinds of centra in the same "phosphor", cannot therefore have anything to do with the accumulation of electrons, only the molecule called the centrum can play a part in that process.

At first there seemed to be some foundation for the supposition that atoms that had passed over from the surrounding material to the centrum would be able to cause storing of electrons. If, however, the result that has begun to appear probable from the more detailed investigations of P. LENARD, *viz.* that the additional substance has no influence upon the intensity nor, *independent* of that circumstance, upon the duration of the bands, will be confirmed, then in the first place the sulphur atoms in the centra are to be taken into account as responsible for the efficient and lengthy storing of electrons. For it is only substances containing sulphur, "phosphors" of the alkaline earths, and zinc sulphide, that show persistent phosphorescence.

Whatever those places in the molecule which we have called centrum may be which have the power of storing electrons, we shall begin by accepting, for trying an explanation, that the electron which is ejected from the metallic atom must in every case reach one of those places before it can be retained. By no means every wave-length of the exciting light seems suitable for this purpose. But not even the most suitable wave-lengths, corresponding with the places of maximum duration in the excitation-distribution, can in consequence of the heat motions of the molecules of the "phosphors" and their atoms that cause the individual centra to simultaneously assume different configurations, be the most suitable for *all* centra *at the same time* for ejecting the electron just in the direction of the place in which it can be stored.

This is the explanation of the circumstance that in "1904" (p. 281, 477, 475) it was found, that in order to develop the afterglow to the full and in the same manner to have good storing of electrons in the lower momentary phase long exposure to, or at least great intensity of the exciting light was necessary. For duration and

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[1]) See also the comprehensive résumé "1909" p. 14. In P. LENARD and SEM SAELAND Ann. d. Phys. 28. (1909). "the surrounding of the centrum" is inadvertently spoken of twice instead of "the surrounding of the metallic atoms" as the place where the electrons are stored.

intensity both increase the number of electrons ejected from the metallic atom (after intermediate returns), and thus also increase the probability that amongst these there will be found some whose direction is the proper one. In the same way, it can be explained that the momentary process, the glow during the illumination without afterglow, in the lower momentary phase is not entirely wanting in the permanent part, for it is just the return of the electrons which have not reached the place where they are stored that causes this glow.

§ 8. *Non-resolution of bands in amorphous substances.* The supposition that in consequence of heat motions the atoms of the centra temporarily assume different configurations also affords an explanation of the broadening and narrowing of the emission bands at high and low temperatures respectively, analogous to that given by P. LENARD in explanation of the broadening of the metal-lines in flame-spectra [1]), and to that given by H. and J. BECQUEREL and H. KAMERLINGH ONNES at the end of their paper for the modification of the positions of the bands of uranyl under the influence of the proximity of other atoms which form with it a molecule. For the different configurations will cause varying changes in the periods of the inherent vibrations of the centra. Without heat motion, at 0° K., each centrum would have a definite time of vibration, invariable with the time. It still depends, however, upon a particular circumstance whether these inherent and time independent vibration periods of all centra (of the same kind) in the whole preparation should in each case be constant, which would be necessary in order to obtain a spectrum consisting of fine lines. For this all centra would have to take up the same position with respect to the neighbouring molecules. For these molecules in a solid substance act with different forces upon the centra according to their distances and orientation, and these must cause different modifications in the period. These equal distances and resulting equal periods for all parts of the preparation independent of time *and* position are to be expected only in crystalline substances such as those investigated by the above-mentioned experimenters, and not in the amorphous "phosphors" investigated by us; this affords an explanation of the fact that even at 14° K. we did not yet see the spectra resolved into lines. What we saw was — according to the image just developed — the overapping of bands consisting of the sharp lines which each centrum gives at very low temperatures, and which by mutual interaction

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[1]) LENARD. Ann. d. Phys. 17 p. 232, 1905.

become less sharply defined. The same remark may be applied to the excitation- and extinction-distributions [1]).

§ 9. *On the upper temperature limits to the excitation of phosphorescence by light, and on the glow of "phosphors" when heated in flames, or when crushed at high temperature.* It seems of importance to us to remark that our demonstration of the existence of phosphorescence, and particularly of the storing of electrons when the temperature sinks to 14° K., also taken in connection with an earlier research [2]), affords a proof that the photoelectric action at low temperatures remains unchanged even down to this very low temperature. This proof was first given by LIENHOP [3]), of the Kiel Laboratory, for temperatures sinking to close upon that of liquid air, from which the validity for all temperatures followed with the highest probability; and for high temperatures MILLIKAN and WINCHESTER[4]) and LADENBURG[5]) have given important direct confirmations. It seems therefore to be beyond doubt that the ejection of electrons by atoms in response to excitation by suitable wave-lengths takes place at all temperatures; and it must, therefore, be explained why the phosphorescence in response to light excitation of the alkaline earths cannot be noticed above a definite high temperature.

It has already been shown that this upper limit to the induction of phosphorescence by light lies lowest in the Ba-"phosphors" and higher in the Sr- and Ca-"phosphors" — *e.g.* in Ba Bi β at 170° C., in Sr Bi β above 300° C., and in Ca Bi β a little above 500° C. — and that it, however, is in no case a sharply defined temperature, but that at high temperatures the excitability by light slowly ceases ("1904" p. 452). The observation has later been added [6]) that it is just in the neighbourhood of this limit of temperature that the "phosphors" begin to conduct electricity. If we assume that this conduction is electrolytic, then it is an obvious deduction that the presence of electrolytic ions prevents the excitation of phosphorescence by light, perhaps through the circumstance that the lost electrons are brought back to the metallic atoms which have been rendered positive by their ejection of electrons under photoelectric

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[1]) The latter according to "1909" should offer the first chance of the resolution into a group of lines.

[2]) P. LENARD and SEM SAELAND. Ann. d. Phys. 28 p. 476 1909.

[3]) A. LIENHOP, Ann. d. Phys. 21 p. 211, 1906.

[4]) MILLIKAN and WINCHESTER, Phil. Mag. July 1907.

[5]) E. LADENBURG, Verh. d. d. phys. Ges. 9 p. 165 1907.

[6]) P. LENARD and SEM SAELAND, Ann. d. Phys. 28 p. 498, 1909.

influence, not as free electrons, but by anions, and it must then be assumed of this kind of neutralisation, that it is not accompanied by emission of light. If the "phosphor" is in a medium containing many free electrons, such as *e. g.* a flame [1]), then it appears to be able to phosphoresce far above that temperature limit; it lights up of its own accord with the phosphorescence colour belonging to this temperature, while light at this temperature is not necessary to induce phosphorescence, and indeed could not do so. To this group of phenomena also belongs that described in "1904" of the "phosphors" lighting up with colours, when after being freshly prepared and still glowing with heat, they are crushed by pressure. In that case there always occurs a burning of the sulphur from the "phosphor" by contact with the air; in fact this glow is seen only where fresh cleavage surfaces formed in the crushing come in contact with the air, and there also small sulphur flames are often to be seen. If too rapid cooling is prevented, addition of oxygen increases the glow. If the "phosphor" is too cold to show the phenomenon of the burning away of the sulphur, then also breakage causes no glow.

On the other hand by holding it on a platinum wire in a flame one can easily keep a piece of "phosphor" — *e. g.* Ca Bi Na or Sr Bi Na — that has not previously been under the influence of light, glowing for a long time with its phosphorescence colour (heat band) at a red heat, and therefore above the temperature limit at which it could be excited by light. The flame must be poor in oxygen if the phenomenon is to last, for as soon as the "phosphor" is deprived of sulphur by burning, the glowing ceases.

We further remark that according to the theory above developed the upper temperature limit of phosphorescence excited by light seems to be a consequence of the electrolytic dissociation of the material of the "phosphor", while its entering the upper momentary phase — which happens for the different bands even of the same "phosphor" at such widely differing temperatures, *e.g.* a little above $-180°$ C. for Ca Bi γ, and at $+450°$ C. for Ca Bi β — can depend only upon processes within the corresponding centrum, that is to say, upon the rendering of electrons from the place where they have been stored in the centrum occurring at temperatures differing widely according to the nature of the centrum.

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[1]) P. Lenard, Ann. d. Phys. 9. p. 649 1902.

P. LENARD, H. KAMERLINGH ONNES and W. E. PAULI. "The behaviour of the phosphorescent sulphides of the alkaline earths at various temperatures, and particularly at very low temperatures."

Fig. 1.

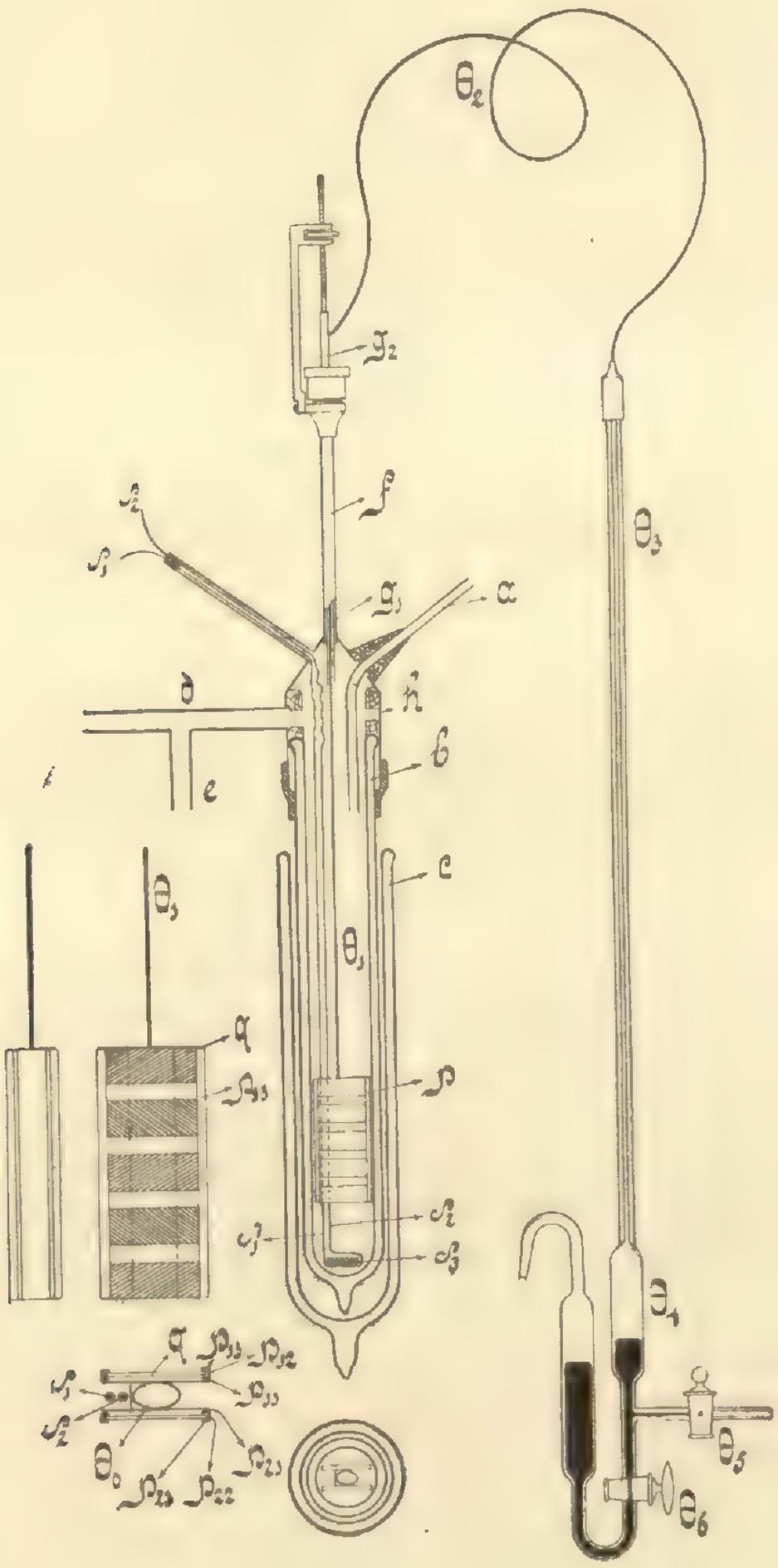
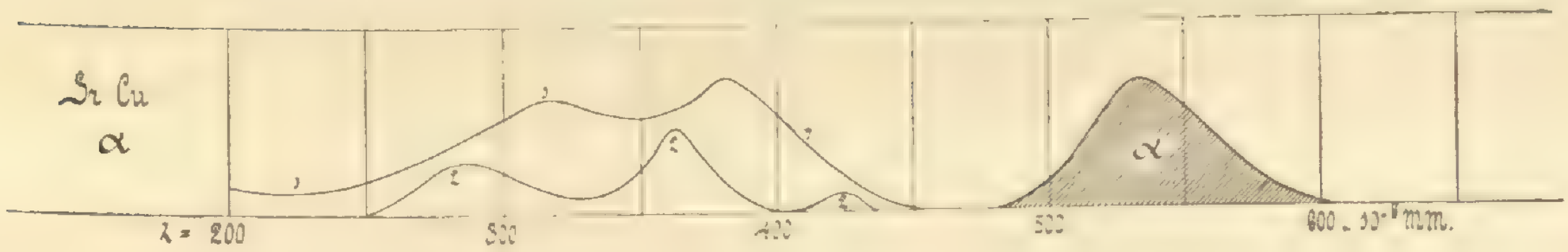


Fig. 2.



1. Excited intensity during illumination (momentary + permanent excitation).
2. Excited intensity in the afterglow (permanent excitation).

Physics. — "*Isotherms of monatomic gases and their binary mixtures. III. Data concerning neon and helium.*" By Prof. H. KAMERLINGH ONNES. Communication N°. 112 from the Physical Laboratory at Leiden.

§ 1. *Critical pressure and triple-point pressure of neon.*

Mr. G. CLAUDE of Paris had with extreme kindness placed at my disposal a considerable quantity of a gas rich in neon which had been obtained by fractional distillation of air, carried out by the Paris Société de l'Air-liquide according to CLAUDE's method. By means of my hydrogen circulation I was in a position to prepare from this a quantity of pure neon quite sufficient for the determination of various physical properties of this gas which, up to the present, has been but little investigated. When the pure neon was first collected in the mercury pump, a characteristic light phenomenon was at once apparent; this phenomenon seemed to have been unknown up till that time, although, as appeared from a paper published shortly afterwards, it had been observed by Prof. COLLIE, who also was indebted to the kindness of Mr. CLAUDE, and had been further investigated by him. The phenomenon is this: — whenever the mercury sinks from the reservoir of the TÖPLER-pump to the lower tube, the meniscus is surrounded at the contact with the glass, by a narrow ring of clear orange light. By this neon can be immediately recognised.

The triple-point pressure and the critical pressure were determined in the following manner. Pure neon was condensed in a glass tube which a steel capillary connected with a manometer tube and a valve through which gas from a reservoir could be admitted. The condensation was easily brought about by immersing in liquid hydrogen the experimental tube, surrounded by a protecting tube that served to lessen heat-conduction and thus to delay the temperature changes in the contents of the tube. Then by allowing the neon to flow quickly into the apparatus it was obtained in the liquid state in the experimental tube. If the protecting tube is kept surrounded by liquid hydrogen, then the experimental tube cools slowly, and after a time the neon begins to solidify. If, on the other hand, the liquid hydrogen surrounding the protecting tube is sucked away the temperature of the experimental tube slowly rises, and if, by means of the valve, the quantity of neon in the tube is well regulated, the meniscus is seen to disappear somewhere in the tube. By allowing the gas to expand through the valve the meniscus can be made to re-appear in the middle of a mist, just as it happens in ordinary determinations of the critical state. When the meniscus disappeared

the critical pressure was read on the manometer tube attached to the experimental tube.

For the triple-point pressure was found 35 c.m., and for the critical pressure 29 atmospheres.

A bath of liquid neon would therefore prove of great advantage in investigations in the neighbourhood of the critical point of hydrogen. I hope to be in a position to make use of such a bath through the further kindness of Mr. CLAUDE, to whom I now express my warmest gratitude for the help already given.

§ 2. *Critical pressure of helium. Lowest temperature reached.*

In the first liquefaction of helium I made use of a very provisional estimation of the critical pressure giving a value of 7 atm. The observation of the vapour-pressure and the density of liquid helium showed that the critical pressure must be much lower and must lie rather between 2 and 3 atmospheres.

If this value had been taken, then a lower value would also have been chosen for the pressure under which the gas to be liquefied was pumped through the regenerator spiral. That there was, in fact, occasion for this was shown by the circumstance that in the actual experiment the expansion pressure was lowered with successful result. Indeed, had account not been taken of a warming effect that was observed and of the knowledge that the JOULE-KELVIN process can give a warming if the pressure is too high even when the temperature is suitabie for the production of liquid the experiment might perhaps have been unsuccessful; and the failure might have been attributed to a very special deviation of helium from the law of corresponding states, and hence to the expansion pressure not being sufficiently high, or to an error in the estimation of the critical temperature.

When, therefore, a further liquefaction of helium was proceeded with [1]) (which had been delayed by various circumstances for a considerable time) the pressure in the regenerator spiral was from the beginning brought to a value of 50 atmospheres. With this the liquefaction proceeded as well as could be desired. To keep the circulation in action once the liquid was formed, it seemed advantageous to bring the pressure in the regenerator-spiral even to the

[1]) In the first repetition of the experiment we did not succeed in preventing the formation of a slight deposit on the hydrogen glass. Had this been the case to the same extent in the first experiment, then there had been every probability that, on account of the first observation of the liquid helium being much more difficult, the experiment had been abandoned as unsuccessful, while as a matter of fact, the apparatus contained liquid helium.

still lower value of 20 atmospheres. Just as has already been remarked, now that the conditions for the liquefaction of helium are known, the repetition of the experiment with the same apparatus and the keeping of 60 c.c. of liquid helium in it for several hours has become a work that offers no serious difficulty, although it necessitates much exact and careful preparation. Now we can attempt a further step and pour off the liquid helium into a cryostat in which various measuring apparatus can be immersed.

In the new experiment I have been successful in still further lowering the temperature of the liquid helium. It was definitely known that in the first experiment the vapour pressure was lowered to 1 c.m., and that the helium was still liquid at that pressure; now I have successfully lowered the pressure to 2.2 m.m. Even at that pressure the helium remains a light mobile liquid. When lowered to this low temperature it shows considerable shrinkage, and, in correspondence with the greater density clearly shows a rise against the wall of the glass [1]). The temperature which is reached by thus strongly cooling the liquid helium may be estimated to lie below 2°.5 K., perhaps at 2° K.

I have tried in the new experiment to measure these extremely low temperatures with the gas thermometer. It must, however, be remarked that when the temperature sinks so low, the thermometer may be used only so long as the pressure of the helium in the thermometer remains lower than the vapour pressure of the liquid helium, and when the latter is 2 m.m. the pressure in the thermometer at that temperature must be still lower, *e.g.* 1 m.m. The pressure at which the thermometer is filled at ordinary temperature is therefore taken not higher than 100 m.m. The difficulties of determinations with this very special gasthermometer in the immediate vicinity of the helium apparatus have not yet been quite overcome, and hence further information concerning the temperature cannot yet be given.

We can, however, already announce a first experimental determination of the critical pressure: it gave 2.75 atmospheres. While an estimate formed from the observations with liquid helium showed that the estimated value on which the liquefaction experiment was based was much too high, it now seems to show a deviation in the other direction, viz. it gives too low a value for the critical pressure.

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[1]) In the former communication it was said that the meniscus at the boiling point stands out against the wall as sharp as a knife. And, indeed, this reproduces in the best way the first impression which the meniscus makes. Proper illumination, however, even under these circumstances, succeeded in making visible an extremely slight rise of the liquid against the wall of the vessel.

Until a cryostat is obtained suitable for such temperatures as here come into account, 2.75 atm. can be regarded only as a provisional value, and it is perhaps better to round it off to 3 atmospheres.

In the meantime the value obtained in connection with more recent determinations which bring the boiling point still closer to 4° K. confirms the estimate of the critical temperature as being at 5° K. But as has already been mentioned, determinations concerning temperatures and densities will be reserved for a later communication.

The determination of the critical pressure took place practically in the same manner as that of neon. It was done in a small pyknometer, a vessel of 0.5 c.c. capacity with a narrow graduated stem, that was placed along with the reservoir of the helium thermometer (see Comm. N°. 108) in that part of the liquefier destined for the reception of the liquid helium; it was attached to a steel capillary and came outside the apparatus just as the capillary of the helium thermometer. This capillary was then connected with a manometer tube, an admission valve, and a reservoir, just as with neon. (See § 1).

According as the level of the liquid helium in the liquefier was more or less lowered [1]), this pyknometer was partially or wholly immersed in the liquid helium, or came wholly above it; helium from the reservoir connected with the pyknometer could be condensed in it, so that the meniscus could be seen in it or in its graduated stem. For density determinations the quantity of helium admitted to the pyknometer was so regulated that the position of the meniscus upon the graduated stem could be accurately determined, while for determinations of the critical pressure the vaporisation was followed while the meniscus was lowering in the pyknometer. The critical pressure was read at the moment when the meniscus when standing half-way up the pyknometer reservoir disappeared, when the helium surrounding it was sufficiently vaporised. The meniscus could be brought back again for a short time by expansion.

The estimate that temperatures below 2°.5 K. have already been reached depends also upon the critical pressure given. An attempt will be made to reach still lower temperatures by causing the helium to evaporate from still lower pressures; for this purpose the apparatus in which the helium is to be cooled by being allowed to evaporate under pressures still lower than 2 m.m., will be surrounded by a layer of liquid helium to protect it from conduction of heat, and to prevent the formation of unmanageable volumes of vapour.

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[1]) This was brought about by allowing insufficiently cooled helium to blow through the regenerator spiral into the helium liquefier.

E R R A T A.

In the Proceedings of the Meeting of May 29, 1909.

p. 88 line 7 from the top: "emission" and "absorption" are to be interchanged.

Fig. 4 Pl. I belonging to the Communication of HENRI and JEAN BECQUEREL and H. KAMERLINGH ONNES: for 511.84 read 511.48.

Fig. 8 Pl II belonging to the same: for 391.26 read 491.26.

(August 31, 1909).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Saturday September 25, 1909.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van Zaterdag 25 September 1909, Dl. XVIII).

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Physics. — "*On the phenomena which occur when, in a ternary system the plaitpoint surface meets the two sheet three-phase surface.*" [1]) By Prof. A. SMITS and E. C. WITSENBURG. (Communicated by Prof. J. D. VAN DER WAALS.)

(First communication).

(Communicated in the meeting of June 26, 1909).

From the behaviour of the system ether-anthraquinone it may be be readily deduced that very peculiar phenomena must occur when to that system is added a third substance which differs from ether in so far that it does not give critical phenomena with anthraquinone in saturated solutions or in other words does not give the critical terminal points p and q.

In this case the peculiar phenomenon of the occurrence of the two critical terminal points p and q, will still at first be noticed on addition of that third substance, but on addition of large quantities of the same it entirely disappears, because the points p and q gradually approach each other and finally coincide.

In order to elucidate the matter in the most simple manner we will make use of the representation in space as given in Fig. 1.

On the posterior plane is drawn the P,T-projection of the system ether-anthraquinone and on the anterior plane that of the system alcohol-anthraquinone so that alcohol is here our third substance which, as shown by the research, complies with the above demands. This was already anticipated as the critical temperature of alcohol is 243°,1 and the melting point of anthraquinone 284°, whereas the solubility of anthraquinone in alcohol is relatively large. Owing to this it is difficult for the plaitpoint curve of this system to come into conflict with the three-phase line.

From the representation in space it will be plainly seen that within the space between the posterior and anterior planes the behaviour of all possible mixtures of ether-alcohol with excess of anthraquinone may be indicated.

For we should remember that between the interrupted three-phase line *epqd* and the continuous three-phase line *ed* extends a two sheet three-phase surface because, in a ternary system, a three-phase equilibrium is a bivariant system in consequence of which three phases may coexist over a series of pressures at a constant temperature.

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[1]) We wish to take this opportunity to express our thanks to the Amsterdam University Union for kindly enabling us to procure a SCHÄFFER and BUDENBERG pump with accessories, thus rendering it possible to carry out this research.

Fig. 1.

This two-sheet three-phase surface of which we will call the lower sheet the solid vapour sheet and the upper sheet the solid-liquid sheet is limited at the lower temperature by the quadruple lines $e\,E$ and $e_1\,E$ for solid ether, solid anthraquinone, liquid and vapour,

respectively solid alcohol, solid anthraquinone, liquid and vapour. Towards higher temperatures the limitation is formed by the line dd' which is a perfectly straight one and of which each point represents the triple point of anthraquinone.

What in a binary system constitutes a line is a plane in a ternary system and consequently there extends between the plaitpoint curves ab and a_1b_1 a plaitpoint surface which is limited towards lower temperatures by the plaitpoint line aa_1 which relates to ether-alcohol mixtures without anthraquinone, and towards a higher temperatures by the line bb_1 of which each point simply represents the critical point of anthraquinone.

From the fact that in the system ether-anthraquinone the plaitpoint curve comes into contact with the three-phase line, whereas this is not the case in the system alcohol-anthraquinone it follows at once that the plaitpoint surface in the ternary system starting from the plaitpoint curve of alcohol-anthraquinone will approach the two-sheet three-phase surface more and more until a junction takes place where the solid-vapour sheet passes continuously into the solid-liquid sheet so that there is formed a plaitpoint line for saturated solutions which runs over the plaitpoint surface and renders a part of it between $p\,p_1\,q_1\,q$ metastable.

This circumstance now causes very peculiar phenomena which may be readily deduced from the accompanying figure in space.

For a small alcohol-concentration a p,t section of the three-phase surface has been indicated in the figure in space, it being assumed that for this mixture there exist also two critical terminal points p_1 and q_1 which represent the points of contact of the three-phase loops $e'p_1$ and $d'q$ with the plaitpoint surface, therefore also with the plaitpoint curve.

Here it should be observed that, as shown plainly from the drawing, the points p_1 and q_1 do not form the extreme limitation of the three-phase hiatus, for these three-phase equilibria may still exist in the supposed mixture above the temperature of p_1 and below that of q_1 so that the temperature-distance of the three-phase loops which is indicated by the vertical tangents is smaller than that of the points p_1 and q_1.

It is plain that when we take larger quantities of alcohol the points p_1 and q_1 and in consequence also the points r_1 and r_2 will approach each other so that the latter will finally coincide, at which moment the liquid branch of the right loop passes continuously into the vapour branch of the left loop, the situation thus becoming as indicated in fig. 2.

We then see that, at this composition, the points p_2 and q_2 are no longer separated from each other by a region of solid-fluid, and that there exists one temperature between that of p_2 and q_2 at which the three-phase region is contracted to one point.

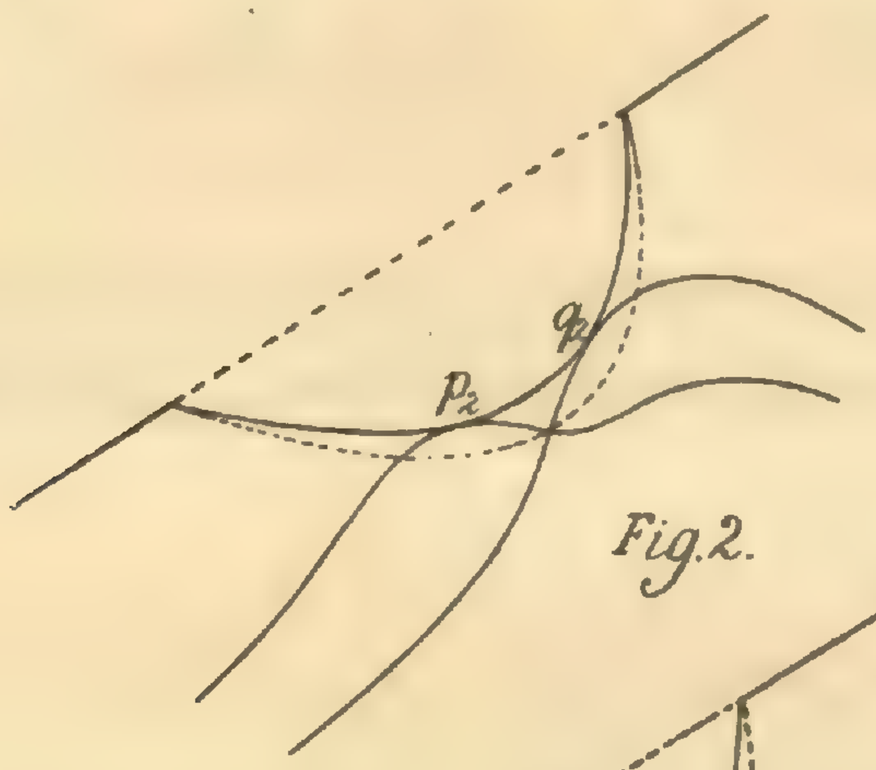

Fig. 2.

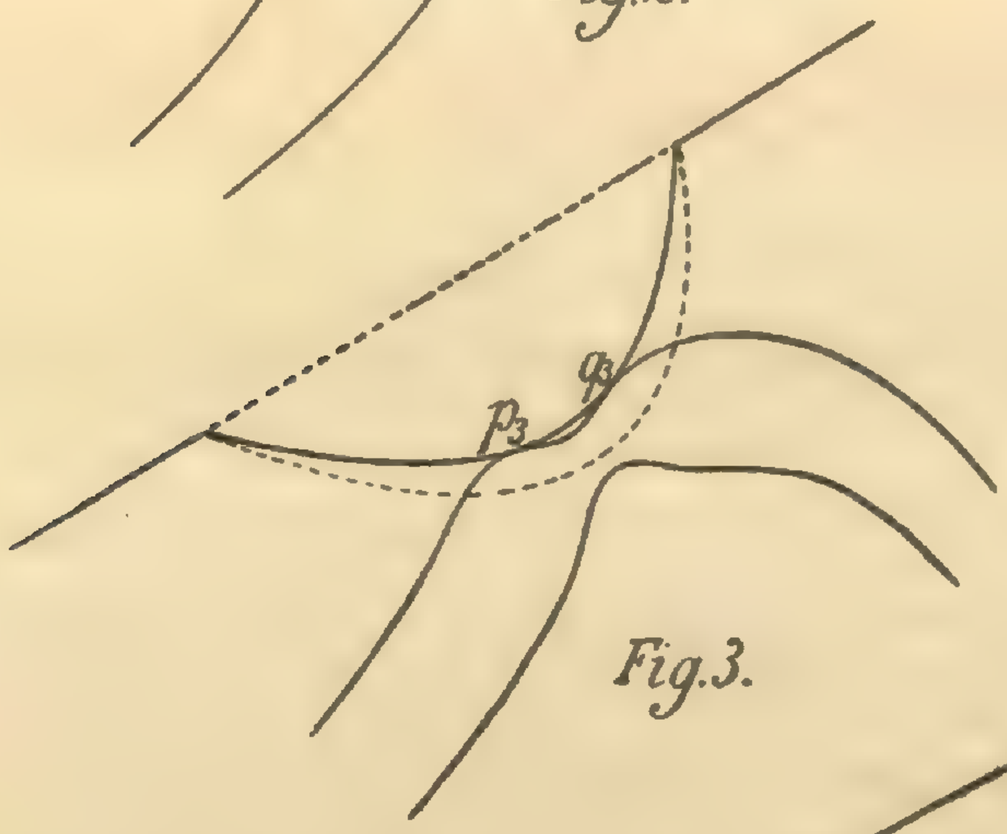

Fig. 3.

Fig. 4.

At a larger alcohol-concentration the situation becomes as represented in fig. 3 and this figure shows the peculiarity of the case even more strikingly than the former, for we now see that although we have passed the inner limitation of the opening, represented by the dotted curve, there are still two critical terminal points, thus without the PT-figure showing a hiatus.

If we now proceed to still larger quantities of alcohol the points p and q approach each other and finally coincide, at which moment there occurs but one critical terminal point on the three-phase area as indicated in fig. 4.

At the end of this first short communication it should be pointed out that the experiment has already proved the correctness of the

above, and that in a following paper a communication will be made as to the experimental confirmation.

At the same time the v,x- and p,x-figures will be considered, which allow a closer investigation of the said remarkable phenomena.

Amsterdam, June 1909. *Inorg. Chem. Lab. University.*

Chemistry. — "*On the compounds of ammonia and water.*" By Prof. A. SMITS and S. POSTMA. (Communicated by Prof. A. F. HOLLEMAN).

(First Communication).

(Communicated in the meeting of June 26, 1909.)

Often it has been asked what happens when ammonia and water come together, but nobody has succeeded as yet in giving an entirely satisfactory answer.

That we are dealing here with a chemical transformation besides the physical process, is shown by the deviations from HENRY's law as well as by the great heat of mixing; but the nature of this chemical transformation is as yet unknown.

If we consider what are the most important facts which we have learned in respect to the solutions of NH_3 in water, these are the following ones:

On dissolution of NH_3 in water the OH'-ion concentration is increased, so that NH_3 dissolved in water behaves as a comparatively weak base.

It further appeared that the solution contains the same NH_4-ions as are split off from ammonium salts, as the electrolytic dissociation in the aqueous NH_3-solution can be reduced not only by increase of the OH'--ion concentration, but also by increase of the NH_4-ion concentration.

The fact that free ammonia is present in an aqueous solution of ammonia was shown by HANTZSCH and SEBALDT [1]); they shook such a solution with solvents for NH_3 such as chloroform from which it appeared that the distribution of NH_3 between water and chloroform depends on the temperature; at higher temperature more NH_3 passes into the chloroform layer.

From this has been drawn the conclusion that on dissolving NH_3 in water we are dealing with a compound whose concentration increases with a decrease in temperature.

The behaviour of the substituted organic ammonia derivatives has

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[1]) Zeitschr. f. phys. Chem. 30, 258 (1899).

led to HANTZSCH and SEBALDT's attempt to render it plausible that this compotind is not NH_4OH, but a hydrate $(NH_3)_m\ (H_2O)_n$ of unknown composition.

ABEGG and RIESENFELD [1]) have felt able to support this idea by demonstrating that the NH_3-pressure of an aqueous ammonia solution increases most on addition of salts which do not yield complex compounds with NH_3, if these salts have themselves a great tendency to form hydrates.

If we consider this and other arguments which have led to the conclusion that in aqueous solution of ammonia hydrates of the same should be present, and not NH_4OH, it appears that the argument is far from convincing and that the conclusion is, therefore, subject to great objections.

When instead of NH_4OH we write $NH_3 . H_2O$, we only mean to say in fact that we are dealing with a compound which does not behave as a base. Now, the solution of NH_3 in water does behave as a base, but this it is endeavoured to explain in the light of the theory of WERNER, by assuming that a part of the free NH_3-molecules will, in consequence of their secondary valencies, unite with the $H^{\cdot}$-ions of the water to cations, the water equilibrium being shifted in such a manner that the OH-ions may appear in large excess.

It is true that the phenomena observed may be explained, even when we deny the existence of NH_4OH,, but for this denial there is no sufficient evidence, while there is not a single phenomenon opposed to the assumption that NH_4OH occurs in the aqueous ammonia solution, which is, on one side, in equilibrium with NH_3 and H_2O and on the other side with the ions $NH_4^{\cdot}$ and OH'.

The research, the first results of which will be communicated here, did not relate to the investigations of previous workers but was simply undertaken to answer the question "what are, apart from the structure, the compounds which may be deposited on cooling from mixtures of NH_3 and H_2O". This question seemed to us an important one, also because from the separation of a compound in solid condition, it may be concluded with a high degree of probability that such compound was previously present in the solution.

The investigation of the melting point line now gave the following result.

In this graphical representation the results of PICKERING's investigations [2]) are indicated by circlets, and thus we notice that, whereas

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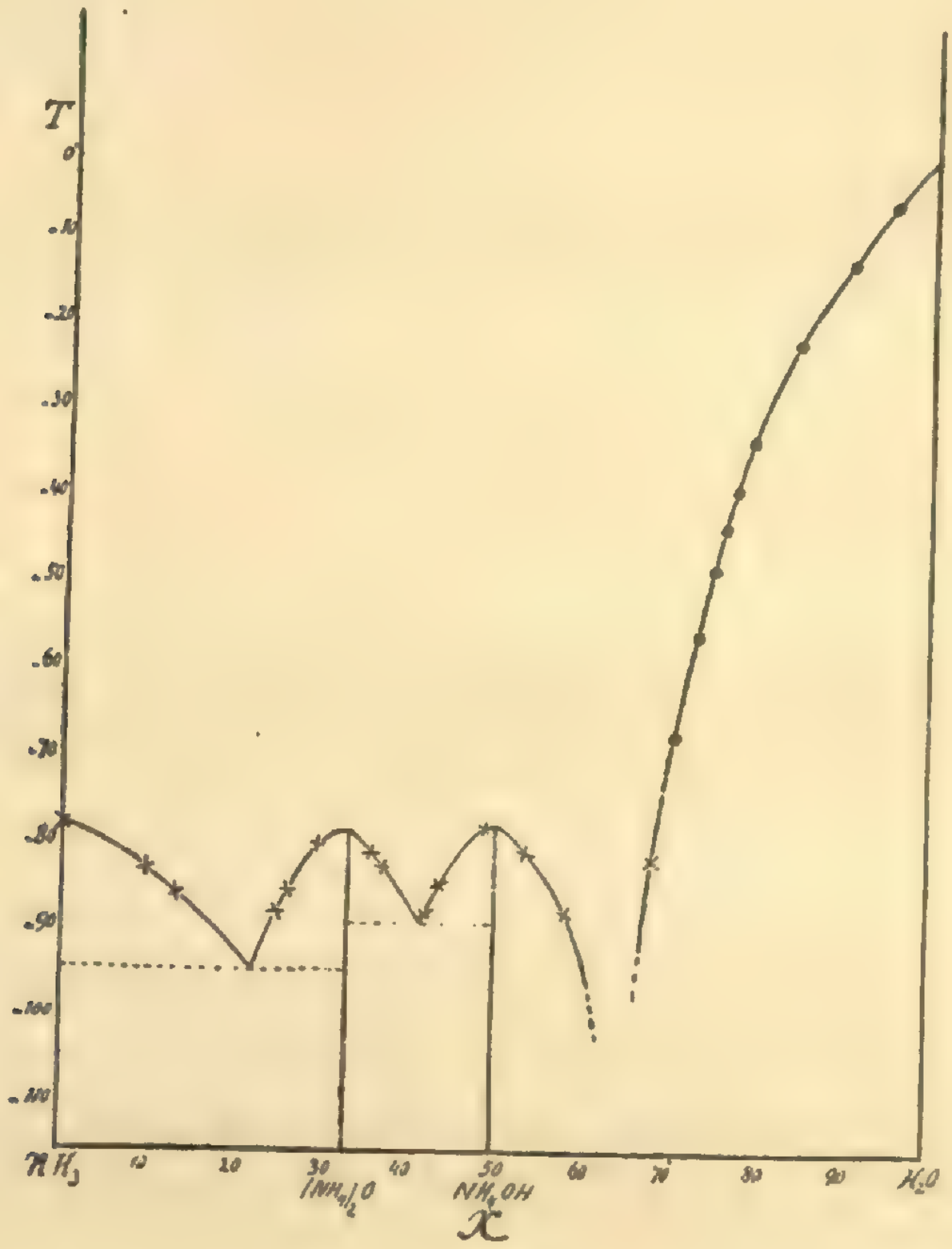
[1]) Zeitschr. f. phys. Chem. **40**. 84, (1902) and **45**. 462 (1903).

[2]) Journ. Chem. Soc. **63**, 141 (1891).

owing to an incomplete investigation, he has found nothing special, two compounds have now been discovered in which NH_3 and H_2O have united in the proportions of 1 : 1 and 2 : 1.

This result is not without significance; for looking at the analogy



between the ammonium salts and those of the alkalis could we expect that NH_4 might combine with the OH-group, and with oxygen in the same proportions as in the case of potassium, and if now, for the reasons stated above, we do not write the compounds found in the form of hydrates but in the form of ammonium salts, we obtain indeed NH_4OH and $(NH_4)_2O$, the first of which melts at — 77° and the second at — 78°.

The communication of further details will be postponed till later on, but here it may yet be mentioned that, as was to be expected, at the water-side and at a low temperature, the investigation was much impeded by the great viscosity of the liquid, so that the eutecticum of H_2O and NH_4OH could not be determined for the present.

This investigation, continued in various directions is also being extended to amines.

Amsterdam June 1909. *Inorg. Chem. Lab. University.*

Physics. — "*An improved semicircular electromagnet*". By Prof. H. E. J. G. DU BOIS. (Communication from the Bosscha-Laboratory).

(Communicated in the meeting of June 26, 1909).

Starting from my theory of the split-ring magnet and its experimental confirmation by H. LEHMANN [1]) I designed a large ring-electromagnet in 1894, which I believe may claim to be considered the prototype of various apparatus since described [2]). Since the general introduction of magnetically efficient cast material it proved feasible to deviate from that simple shape and to redesign it (in 1898) as a so-called semicircular magnet [3]); this was much lighter and handier in many respects and a great number of this type have since been in use in laboratories.

Within a cylindrical "interferric" space of 1 mm. length and 6 mm. diameter their field amounts to 38—40, averaging rather more than 39 kilogauss, in consequence of the somewhat variable properties of cast material. After some ten years of varied experience it now again appears desirable — as indeed it does for most kinds of instruments — to further revise the design with a view to practical use. As the resources of a well-equipped experimenter will in many cases enable him to do good work within narrower interferric spaces, I could attempt to increase the field up to 50 kilogauss without exceeding a total weight of about 300 kg. — distributed over three equally heavy pieces; the apparatus remaining sufficiently easy to handle and carry about in laboratories.

In the meantime Prof. PIERRE WEISS described an electromagnet of great power [4]), which uses up to 18 kilowatt and weighs about 1300 kg.; by this, however, probably only a few privileged laboratories are benefited, and it appears, moreover, less adapted for the important class of modern magneto-optic investigations on account of the long path of the rays of about 125 cm. I could, however, partly, avail myself of his theoretical considerations and results as well as of some details of his design.

Fig. 1 is reproduced at a tenth of the real size. The base part

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[1]) H. DU BOIS, Wied. Ann. **46**, p. 485, 1892. — H. LEHMANN, Wied. Ann. **48**, p. 406, 1893.

[2]) H. DU BOIS, Wied. Ann. **51**, p. 537, 1894.

[3]) H. DU BOIS, Verh. Berl. physik. Ges. **17**, p. 99, 1898; Zeitschr. für Instr.kunde **19**, p. 357, 1899; Ann. der Physik. (4) **1**, p. 199, 1900.

[4]) P. WEISS, l'Eclairage électr. **15**, p. 481, 1898; Journ. de Phys. (4) **6**, p. 353, 1907. — G. ZINDEL, Electrotechn. Zeitschr. **30**, p. 446, 1909.

rests on a broad ball frame with a divided circle, so that the apparatus may easily revolve round a vertical axis; it can also be fixed by means of 3 set-screws. The adjusting screws on the left-hand side are useful for the vertical position of the magnetic axis occasionally required (see fig. 2); the hand-wheel allows gradual displacement of the left-hand core. In practice it is extremely useful and convenient to divide the magnetic circuit by the upper horizontal plane of the base part of the apparatus, so that each of the two cores may slide as well as revolve on it. Each core corresponds to about one third of a toroid and its lower end terminates in a cast flange. Its vertical circular end-planes have a diameter of 93 mm.; towards the bottom this gradually increases.

The investigation of the FARADAY and ZEEMAN effects carried out in this laboratory enforced the condition of great intensity of light, which may be fulfilled by concentration of the beam of rays in the interpolar space, so that it may form a strongly diverging double cone of light. Evidently the borings in the iron must correspond with this, from which the further condition of a short path of the rays immediately follows, as the width of the boring is naturally limited. Conical holes were bored 1 : 5 of a length of only 15 cm.; one of the iron filling-plugs to be used in non-optical experiments is represented in fig. 1; the length of the path of the rays within the iron is restricted to these borings themselves by the peculiar design of angular flanges, which appears sufficiently clearly in the figure. These flanges are provided with regularly spaced nuts, to which optical and other appliances may be screwed, so as to lie just at the mouth of the boring. The polar ends of the cores are surrounded by a thin copper tube for water circulation; 50 litres of water per hour works sufficiently well; moreover an inner circulation may be arranged within the borings; besides the pole-shoes may be provided with external cooling-spirals, such as are clinically used for cooling parts of the human body [1]).

The external shape of the coils appears from fig. 1; the polar flanges are conically truncated in order to make the interpolar space more accessible. When, however, the highest degree of magnetisation is required, two extra detached polar coils may be fixed around the pole-shoes; in accordance with a well-known rule of KIRCHHOFF's the purpose of this is to saturate the polar pieces as far as possible; this principle was, moreover, already applied by H. LEHMANN in his investigation (loc. cit. p. 424). We then obtain 100 (120) kiloampère-

1) Cooling the wire-coils themselves, e.g. by circulating oil is certainly desirable, but rather elaborate and expensive.

turns with 5(6) kilowatts, and the heat generated by the current amounts to 1,2 (1,45) kilocalories per second.

With this apparatus the usual disk-poles and plane polar pieces are supplied to obtain extensive uniform fields. For stronger fields the diameter is reduced in the first place from 93 to 43 mm. by means of "pole-shoes" P of special broken truncated shape (fig. 3 real size). They consist of annealed Swedish laminated iron; their most efficient form was again empirically determined and hardly deviates from the shape used until now [1]). Interpolar pieces of different kinds may then be placed between the two smaller circular end-planes, as required for experiments.

The comparatively small diameter of 43 mm. makes it easy and desirable for every laboratory itself to have such pole-tops Q turned out of Swedish iron, as are best adapted to any special purpose. Their contribution to the total field amounts to 80 to 90 percent, and thus far exceeds that of the pole-shoes, which as a rule only contribute 10 to 20 percent; the influence of the conic surface-elements increases very much as they lie nearer the extremities, where they should be carefully shaped and centred; this purpose is effected by special centred distance-pieces of unmagnetic material. Thus in course of time polar pieces were made for cryo- and pyromagnetic experiments, for micro-magnetic and magneto-optical investigation (FARADAY, KERR and ZEEMAN-effect), for the torsion-balance and magneto-hydrostatic method and that of the isthmus for magnetic stress determinations. A fuller description of these various arrangements would claim more space than we can afford to give it here; it will appear elsewhere with a discussion of the general principles for the design of such intrapolar apparatus, as are adapted to various purposes.

In the conic borings (see fig. 3) split cores or solid filling-plugs may be introduced according to the nature of the experiments. With the pole-tops reproduced in fig. 3, whose end-planes have a diameter of 3,6 mm. the strongest field was empirically obtained through calculation, measurement and mechanical shaping proceeding simultaneously. On the other hand the dotted lines represent the well-known theoretical optimum form consisting of a cone with a vertex semi-angle of 54°44′, and part of the "isthmus". The theory must start from the assumption of absolute saturation in the direction of the axis, which can never really be fulfilled; hence the actual deviation,

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[1]) Since 1901 slightly rounded pole-shoes were supplied with large half-ring electro-magnets. No pole-shoes were required for the smaller ones, and it proved useful, on the contrary, to provide their pole-tops with a bevilled edge, which peripherically increases the effective conic surface. (see fig. 4).

though it is rather slight. It proved useful again to let the vertex angle increase from the centre towards the edge, from about 56° to 59°; instead of by a broken line, this transition can also take place by means of an arc of 3°, and corresponding — rather large — radius; the surface of revolution then becomes somewhat toroidal.

A great number of field measurements with a very small test-coil were first made in order to gradually approximate towards the optimum form, to which, however, rather a "flat maximum" corresponds, which leaves the designer a certain margin. The final results are contained in the following table.

End planes: distance (mm.)	6 mm. Field (kgs)	3.6 mm. Field (kgs)	
0	50 (extr.)	55 (extr.)	
0,5	48.7	52.2	(46)
0,9	—	**50**	—
1,0	**47**	—	(42.5)
1,5	45.3	46.5	(39)
2,0	43.7	44.5	—

These fields were obtained with 124 kiloampère-turns (with extra polar coils); the first limiting value has been linearly extrapolated and is of some theoretical interest; for some experiments intervals below 0,5 mm. are already sufficient. I have placed between parentheses the field values obtained by screwing the same polar pieces into the old ring-electro magnet, 108 kiloampère-turns (without extra polar coils) being used in this case.

In fig. 2 a smaller type is represented in an erect position at $^1/_5$ of the real size. This is essentially a simplified linear reduction to about $^2/_5$; it is well-known that small electro-magnets are relatively much more efficient than larger ones, though it is more difficult to spare sufficient space for the ampère-turns required in the same linear ratio. The conic pole-tops with their bevilled edges — as in fig. 4 — are cast in one piece with the cores themselves, and properly finished on the lathe. This apparatus weighs but 33 kg.; with 24 kiloampère-turns — only $^1/_5$ of the above-mentioned value — the following approximate results were obtained:

End planes: distance (mm.)	6.7 mm. Field (kgs)	3.6 mm. Field (kgs)
0	36 (extr.)	40 (extr.)
0,5	34	37
1,0	32	34
1,5	30	31

I wish to express my great indebtedness to Prof. KŌTARŌ HONDA for his kind assistance in carrying out the necessary field measurements.

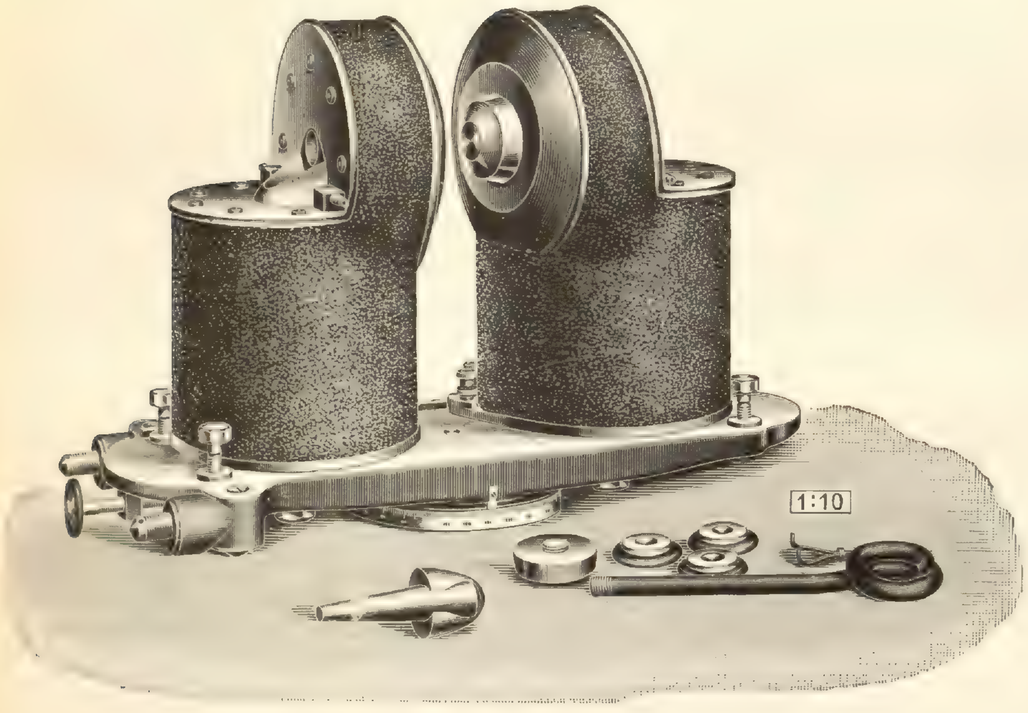

Fig. 1.

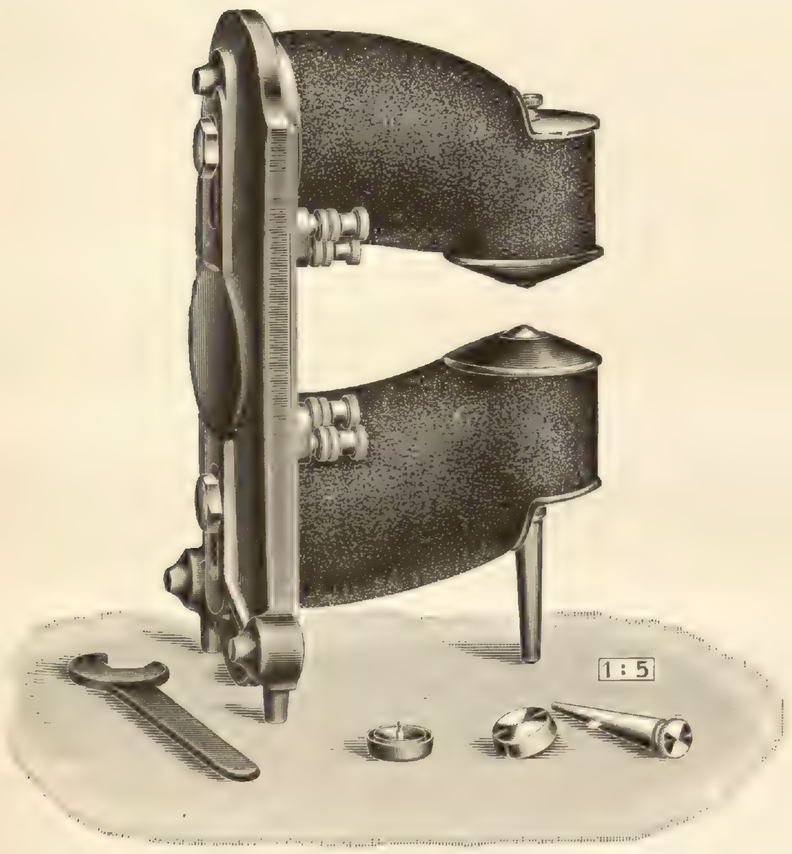

Fig. 2.

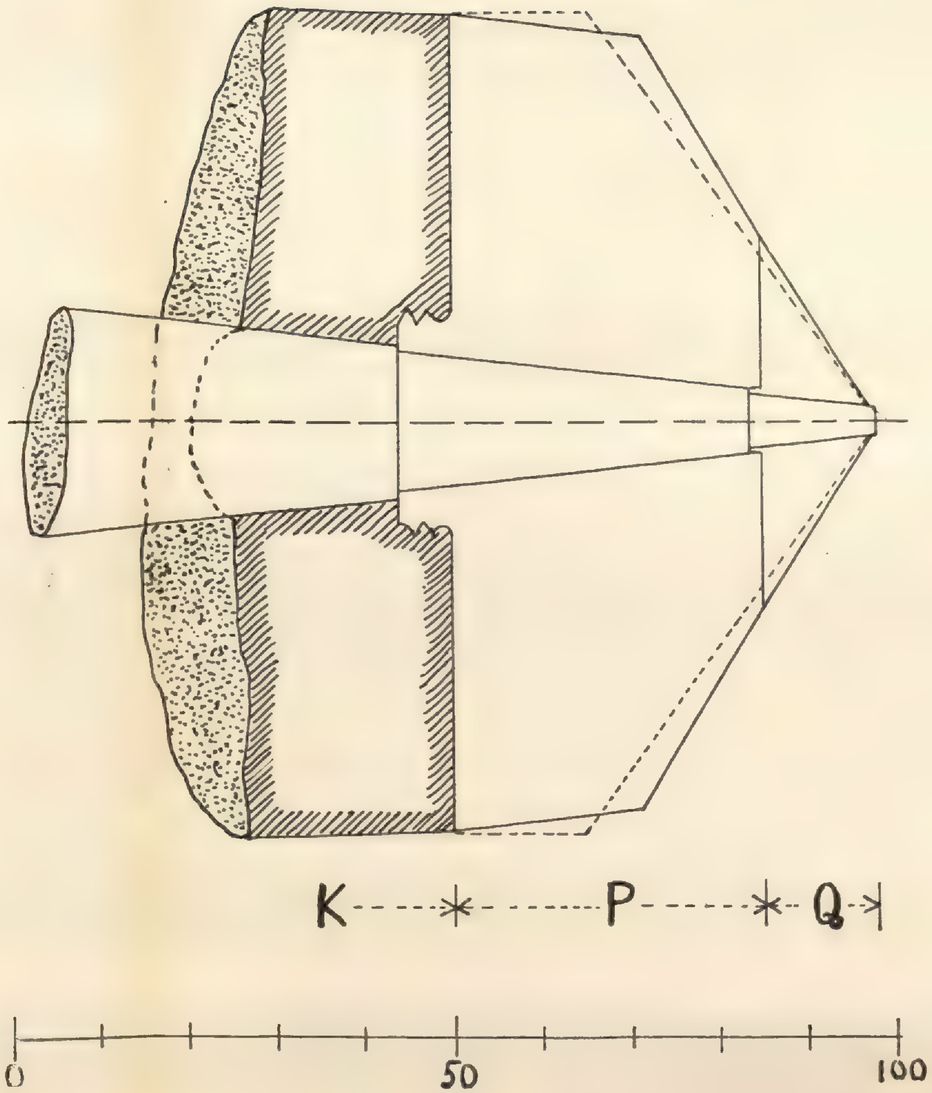

Fig. 3.

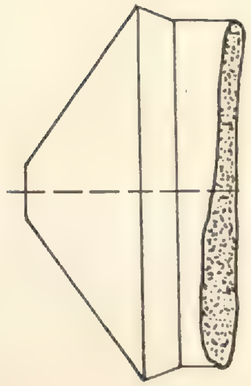

Fig. 4.

Botany. - *"The physiological significance of certain glucosides."* By Dr. TH. WEEVERS. (Communicated by Prof. F. A. F. C. WENT).

In the Proceedings of the ordinary meeting of the Mathematical and Physical Section of September 27th 1902 there appeared an account of some investigations on glucosides in relation to plant metabolism, and of this the present paper is to be considered a continuation.

It was shown at the time that both salicin from Salix purpurea L. and the glucosides from the seeds of Aesculus Hippocastanum L. are reserve materials; the former substance is used in the opening of the buds, the latter in germination.

At the opening of the buds of Salix purpurea saligenol[1]) appears, which points to a hydrolysis of salicin; the quantity found was, however, so small, that the actual aromatic product must be another substance, and as such I regarded catechol, which can be demonstrated in all parts of the plant. When the salicin diminishes, the catechol increases and in various cases the change in the amount was found to be roughly proportional to the molecular weights. These quantitative experiments induced me to suppose that catechol, after hydrolysis of the salicin and transport to other parts of the glucose thus formed, remains localised in the cell and again combines to form salicin with glucose, which is brought from elsewhere or has been formed on the spot by assimilation. Thus PFEFFER's hypothesis would be confirmed, that the compounds of benzene derivatives with carbohydrates serve to form substances not easily diffusible; in the present case catechol would form with the transport substance, glucose, the reserve substance salicin.

In the later investigation the validity of this supposition was tested on another glucoside, namely arbutin, which accompanied by small quantities of methylarbutin, occurs in numerous Ericaceae. Similarly the occurrence of fairly large quantities of hydroquinone, the aromatic product of arbutinhydrolysis is recorded for these Ericaceae. Experiments with Vaccinium vitis idaea L. showed, however, that the latter statement is not quite correct; generally the amount of free hydroquinone is very small, and in autumn it is even zero; it increases, however, considerably during the formation of the young shoots in spring.

Arbutin is rapidly hydrolysed by an enzyme, which I obtained from Vaccinium vitis idaea, so that only those quantitative deter-

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[1]) Saligenol = salicylalcohol; catechol = orthodioxybenzene.

minations give good results, in which the parts have been killed by boiling water or better by boiling alcohol.

The quantitative estimation takes place as follows: the parts of the plants are killed by boiling alcohol, and are then extracted with warm water; the extract is treated with lead acetate, of which the excess is removed with sodium phosphate. The free hydroquinone is obtained in almost colourless crystals by extracting the liquid with ether. After extraction the aqueous solution is boiled for one hour with dilute hydrochloric acid, to hydrolyse the arbutin completely, and the hydroquinone formed is then again extracted by shaking with ether.

In case the amount of hydroquinone was small, the estimation was carried out in the same way as was indicated for catechol in the former paper, viz. by comparing the sublimates with those of a standard solution. If the quantity of hydroquinone was more than a few milligrams, the estimation was carried out by titration with FEHLING'S solution according to ALLIHN'S method, after a table of the amounts of copper, reduced by pure weighed-out hydroquinone, had been prepared as a basis of calculation.

The isolation of methylhydroquinone was carried out by extraction of the ether residue with benzene, in which methylhydroquinone is soluble, but not hydroquinone itself. The amount of methylarbutin and of methylhydroquinone, however, proved to be small, so that no considerable error was made in calculating everything as arbutin and hydroquinone. The arbutin content showed considerable individual variation and as one specimen was too small for an experiment, the experiments were carried out with a large number of plants from the same spot. The leaves are small, so that it was impossible to trace the variations between day and night by the method of cutting the leaves in half; I therefore only determined the changes during the opening of the buds.

Arbutin plays the part of a reserve material, which in Vaccinium vitis idaea, an evergreen shrub, is principally deposited in the leaves, and there increases in amount during the winter until the opening of the buds in May. Then the amount varies from 2—3 % for plants in the shade, to about 5 % for plants in the sun [1]).

Before the opening of the buds no hydroquinone, or merely a trace, is present in the leaves, and the same applies to the stems and roots, which latter at this period also contain but traces of arbutin. With the development of the young shoots from the buds

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[1]) All values calculated for dry weight.

the amount of arbutin diminishes progressively, and that of hydroquinone first increases, but afterwards quickly diminishes when assimilation begins in the young shoots. On comparison of the amount of arbutin in a number of plants (May 3rd) before the opening of the buds, with that after the opening (May 24th), taking old and young parts together, 22—32 % of the total was found to have disappeared.

This glucoside is therefore also used in the opening of the buds and undergoes before use a fermentative hydrolysis, as a result of which hydroquinone shows itself in the tissues. The amount of hydroquinone is, however, much smaller than one might expect from the amount of arbutin which has disappeared (for 100 milligrams of arbutin, for instance, 6 milligrams of hydroquinone). Thus a part of the hydroquinone appears here to be directly worked up in metabolism, and the hydroquinone, which is present in the opening buds, also disappears again rapidly as soon as assimilation becomes vigorous. I therefore found only traces of arbutin in the second budding of some plants in August, which is nevertheless also accompanied by arbutinhydrolysis.

During the opening of the buds I often found that no arbutin occurs in the stem, a further proof that the glucoside is not transported as such.

In the buds of the pear-tree Rivière and Bailhache [1]) have demonstrated hydroquinone, and this induced me to search for a hydroquinone-glucoside in this case also. Such a glucoside was indeed found to exist and the amount of combined hydroquinone was again much greater than that in the free state. The enzyme, which here I was also able to prepare from the young shoots, rapidly brings about the hydrolysis of the hydroquinone-glucoside [2]), so that it is necessary to kill the parts by boiling alcohol. Then one finds in the adult leaves 0.01—0.03 %, in the young shoots 0.3 % of hydroquinone. In this case methylhydroquinone and methylarbutin are absent.

The glucoside is found in the buds, leaves, wood and bark of the branches and also in the roots, but in the bark only. The amount in the root-bark is very small (0.05 %); that in the wood of the

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1) G. Rivière et G. Bailhache. De la présence de l'hydroquinone dans le Poirier. C. R. de l'Acad. de Sc. Paris CXXXIX. 1904.

2) I could not obtain the glucoside in a crystalline form, so that I could not determine with certainty from the physical constants, whether it is identical with arbutin; the products of hydrolysis, hydroquinone and glucose, which I found, make the identity very probable, however.

branches is less than that in the bark. The quantitative determinations, which took place in the same way as in Vaccinium, were carried out on a single tree of the variety Bonne Louise d'Avranches.

The glucoside is formed in the leaves, and the formation begins in the young shoots as soon as they themselves begin to assimilate.

On comparison of the amounts in the halves of a leaf in the morning and in the evening, the glucoside is found to increase during the day and to diminish during the night. This diminution during the night is again accompanied by an increase of hydroquinone and the amounts are in the proportion of 100 : 18 [1]), whereas the molecular weights are in the proportion of 100 : 40. A portion of the hydroquinone, which has been liberated in the nocturnal hydrolysis thus appears to be directly transformed in metabolism, or else we must assume, that a part of the arbutin is transported as such.

During the summer the amount of arbutin increases in the bark, from 1.5 % to 4.5 %, so that we may safely infer, that the arbutin is deposited there in order to be used up in the formation of new shoots in the spring. At that season the amount in the bark diminishes greatly (about 70 % of the total) and increases less markedly in the young shoots, so that the total diminution from March 30th to May 2nd amounted to about 27 %. During this process the amount of free hydroquinone is greatly increased by fermentative arbutin hydrolysis and from April 26th to May 2nd, for instance, the decrease of arbutin was to the increase of hydroquinone in the proportion of 100 : 40, exactly that of the molecular weights; in other words the whole of the aromatic product remains localized in the cells [2]). Afterwards, when assimilation has begun in May, hydroquinone is rapidly worked up and arbutin is formed at its expense. Hence we find PFEFFER's hypothesis once more confirmed: hydroquinone combines with the transport substance glucose, with formation of the reserve material arbutin.

In the case of Salix purpurea I found in 1902 that by slow dessication 25 % of the salicin disappeared from the bark. This pointed to the presence of a salicin-splitting enzyme, but at the time I was not able to demonstrate it. On continuing the experiments

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[1]) In Salix purpurea also the relation of salicin decrease to catechol increase in day and night experiments differed more from the theoretical than in the case of the opening of buds of branches placed in water.

[2]) Most of the hydroquinone formed, was in the young shoots; this might therefore be interpreted as an indication, that a transport of arbutin takes place in this case.

I soon obtained better results: bark from the stem was placed in 96 % alcohol immediately after the pealing of the branches; the alcohol was allowed to evaporate slowly in the air and after some days the liquid which was only feebly alcoholic, was filtered. On the addition of salicin 9 % was hydrolysed after 24 hours, after 48 hours 17 %, after 96 hours again only 9 %, measured by the glucose formed.

Young shoots were also ground in a mortar and pressed through fine plancton gauze, the juice was treated with an excess of 96 % alcohol; a white precipitate was thus formed, which was filtered off, washed with alcohol and finally dissolved in water. The colourless slightly opalescent liquid, thus obtained, did not reduce FEHLING's solution, did not contain saligenol or catechol and hydrolysed on the addition of salicin after 24 hours 8%, after 48 hours at 40° 28%, after 72 hours 46%. The extent of the hydrolysis was measured by estimating the glucose formed; in the ethereal extract saligenol could be demonstrated [1]). From the young shoots a mixture of enzymes had thus been obtained, among which was an enzyme which splits salicin into glucose and saligenol [2]). I obtained the same enzyme from Populus canadensis Mchs.

The salicase is not identical with emulsin, nor with amygdalase, for it does not bring about the decomposition of amygdalin.

The above mentioned experiment with the weakly alcoholic extract of the bark, pointed to a reversible action of the enzyme, as did also some other experiments. For when I dissolved 200 mgs. of glucose and 100 mgs. of saligenol in 2 cc. of water and added salicase (with thymol as antiseptic), the reducing power was diminished by 5% after 2 days at 40°, which diminution might be explained by the possible formation of salicin [3]). I had not, however, sufficient material to make this out with certainty.

The discovery of this salicase induced me to investigate once more the occurrence of saligenol in Salix purpurea, as formerly I had only been able to find minute traces. For this purpose I compared in the

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1) Recognized by allowing the sublimate of the ethereal extract to crystallize from aqueous solution, and by the reactions with cupric acetate and with ferric chloride.

2) When I had already completed my investigation, my attention was drawn to a paper by W. SIGMUND (Sitz. ber. Akad. d. Wiss. Wien. CXVII. Bd IX 1909) who obtained from other species of Salix and of Populus an enzyme, called by him salicase, which hydrolyses salicin, and from Calluna vulgaris and Vaccinium myrtillus an arbutase, which hydrolyses arbutin.

3) After boiling with dilute hydrochloric acid the reducing power was again as great as before.

same shrubs at different dates the quantities of catechol and of saligenol in the young shoots. The quantitative estimation of the latter substance was carried out in the same way as that of hydroquinone and of catechol, by comparison of the sublimates with that of a standard solution; the separation of the two benzene derivatives was achieved by complete precipitation of the catechol by lead acetate.

Per 200 young shoots I found	23	April	34	mgr.	catechol	—	saligenol
	28	,,	26	,,	,,	—	,,
	2	May	9	,,	,,	—	,,
	5	,,	1	,,	,,	—	,,
	8	,,	2	,,	,,	7 mgr	,,
	16	,,	1	,,	,,	24 ,,	,,
	28	,,	35	,,	,,	— ,,	,,

Neither at this time, nor at any other time saligenol can be detected in the bark, and in the young shoots it only occurs for a few days exactly as in the case of the methylsalicylate during the opening of the buds of the beech [1]).

This absence of saligenol is not due to a lack of salicase, for in the bark of the branches, where the salicin diminishes greatly, and in the young shoots, where salicin is used, this enzyme was always present. The saligenol must therefore be directly transformed, and as I have shown before catechol is the probable end-product of this transformation.

The possibility naturally suggested itself of obtaining from Salix purpurea an enzym which should convert saligenol into catechol.

The above mentioned mixture of enzymes readily splits off molecular oxygen from hydrogen peroxide, and therefore contains catalase [2]), but in addition it contains two other oxydases, which differ from laccase and from the system hydrogenperoxide-peroxydase and tyrosine, both on account of their behaviour towards the cresols and because

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[1]) Compare Onderzoekingen over Glukosiden: Diss. Amsterdam 1902. Th. Weevers.

[2]) An attempted separation of catalase and catecholase by heating was unsuccessful, and it might be argued that it is here unnecessary to postulate the presence of a separate catalase; hitherto the splitting off of molecular oxygen, however, has always been regarded as characteristic of catalase and conversely the known vegetable catalases do not give the reactions with catechol and saligenol; for the above physiological considerations this is, moreover, of no importance.

The crude enzyme from Salix purpurea contains no manganese and acts on catechol both in a feebly alkaline and a feebly acidic medium (litmus and rosolic acid as indicators). By heating to 100° C. the enzyme is at once destroyed. The views of Dony Hénault Bull. Ac. Roy. Belgique 1909 are therefore not applicable to this case, nor those of Euler Bolin, Zeitschr. phys. Chem. 1908.

of the absence of the guiacum-blue coloration after addition of H_2O_2. They were named after their typical reactions saligenolase and catecholase; both gave an almost colourless, somewhat turbid solution.

They may be separated by heat, for after heating to 85° C., the saligenolase had been destroyed, but not the catecholase. Catecholase oxydises catechol: a catechol solution which by itself remains unchanged for days, changes at once after the addition of catecholase through green to black and after 5 minutes there is a definite black precipitate; saligenol is not changed by catecholase.

Both oxydases together evidently form from saligenol, although more slowly, the same product as the catecholase forms from catechol. The most obvious hypothesis is therefore to assume that *saligenolase forms catechol from saligenol.*

In any case this oxidation of saligenol (salicyl-alcohol) is quite different from that in the laboratory, where salicylic acid is always the final product [1]).

In the living plant this black substance [2]) never appears, but it only occurs in necrobiosis; therefore the most attractive hypothesis seems to me that catecholase and catechol are separated from each other in the cells but that this is not so with saligenolase and saligenol, so that the latter enzyme can act and can form catechol, which then cannot be decomposed by catecholase, as is the case in necrobiosis and in the above mentioned experiments.

In the former paper I investigated the changes of salicin and catechol in branches, with buds opening, when placed in water in the dark, and now I have done the same with branches budding while still attached to the plant. The objection in this case is, that the branches do not form a separate whole, so that influx and exit is possible, while assimilation very soon sets in. For all these reasons one may expect the observed values for the relation of the salicin disappeared to the catechol formed to differ more from those calculated by theory. I thus found, for instance, an increase of 94 mg. catechol accompanied by a decrease of 457 mg. salicin, i.e. a proportion of 21:100, while the molecular weights are in the proportion 38:100. I also found however, that in the opening of the buds the amount of populin increases at the expense of salicin, so that the diminution in the glucoside which furnishes catechol on hydrolysis was placed at too high a value.

I also repeated on a larger scale the earlier experiments with

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[1]) Neither catecholase nor saligenolase have any action on salicylic acid.

[2]) Perhaps this black colouringmatter is the same as that formed from homogentisic acid under the influence of alkali.

etiolated budding branches and with bisected leaves (the halves of the leaf being compared in the evening and in the morning). In etiolated budding the catechol increase and salicin decrease were in the ratio 36 : 100; in leaves the nocturnal increase of catechol was to the decrease of salicin in the ratio 31 : 100 (for 2000 leaves), which agrees sufficiently well with the hypothesis.

It is not until the salicin content of the bark becomes great (6 à 7 %) and equals that of the leaves, that the salicin ceases to diminish in the leaves overnight and the catechol no longer increases, the glucose transport of the hydrolysed salicin then seems to have stopped. Whether the salicin concentration influences the transport, or whether the concentration gradient of the glucose does so, remains for further investigation.

In order to determine also the populin present in the bark of Salix purpurea I used a press-juice obtained from Aspergillus niger, which was precipitated by an excess of alcohol. The precipitate contained a mixture of various enzymes, among which is one which completely hydrolyses populin, as was shown by experiments on the pure glucoside.

In addition, the mixture of enzymes also contained emulsin, invertin and maltase; in order to utilize this mixture for the quantitative determination of populin the increase of reducing sugar after the action of the Aspergillus enzyme was to be diminished with that, obtained after the action of emulsin, invertin [1]) and maltase [2]).

As might be imagined the method only gives useful results, when the populin is present in large quantity, for in the glucose-values found for populin all the errors of the other determinations accumulate, and these errors can never be completely excluded in the case of hydrolysis by enzymes.

The method cannot therefore be applied to Salix purpurea, where the populin is quantitatively unimportant. It was found, however, that populin is formed in large quantity in the normal young shoots but is, on the other hand, wholly absent from etiolated shoots; this is the reason why in the etiolated budding the ratio of salicin to catechol agrees so much more closely with that which might be expected theoretically.

I also attempted to determine populin quantitatively in Populus species, but did not obtain good results, because the populin content is too small in the species I have hitherto examined, viz: P. alba L..

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1) Salix purpurea contains small quantities of saccharose in the leaves and bark.

2) The extract had been obtained from the parts with warm water and had been treated with lead acetate, so that it did not contain any starch or dextrins.

P. canescens Sm., P. monilifera Ait. and P. tremula L. I found here, as in the case of Salix that the young etiolated shoots contain no populin and the normal ones a fair amount. In all the species investigated, salicin is present and also catechol; for one of these plants, P. monilifera Ait. the changes of the two substances during the formation of the young shoots were studied.

In the opening of buds on the tree as well as in etiolated budding of branches placed in water, salicin decreases, catechol increases; the quantities are in the ratio 100 : 66 and 100 : 64. The increase in catechol is therefore greater than might be expected from the decrease in the salicin, and this I think must be ascribed to the diminution in the populin (benzoylsalicin).

I obtained from this Populus, in the manner described above, an enzyme, populase, which splits off benzoic acid from populin, so that the formation of catechol as end-product of populinhydrolysis is indeed very probable [1]).

In Populus monilifera and also in P. tremula salicin increases in the leaves during the day and decreases during the night, just as in the case of Salix; a quantitative determination of the catechol was however impossible on account of the resin present.

Catalase is present in the young shoots and also catecholase; the demonstration of the presence of saligenolase proved difficult as the enzyme mixture is not so pure and becomes darker fairly rapidly when exposed to air.

In the species of Populus examined I found considerable quantities of saccharose, which there plays the part of a reserve material and is present in the leaves, bark and wood (Populus monilifera Ait. and P. tremula L.). In the opening of the buds a large portion is used and the amount in the bark than falls from 4 % to 1 %; the experiments with bisected leaves also point to a behaviour similar to that observed by Brown and Morris [2]) in Tropaeolum. During the night 32 % and 35 % of the total disappeared from the leaves. Like so many other plant organs the young shoots accordingly also contain invertin. For further details reference may be made to the publication about to appear in the Recueil des Trav. Bot. Néerl.

Amersfoort, September 1909.

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[1]) With the mixture of enzymes from Salix purpurea I could not obtain any hydrolysis of populin.

[2]) Brown and Morris. Journ. of the Chem. Society. 1893.

Anatomy. — *"Surface and structure of the cortex of a microcephalic idiot."* By Dr. C. T. VAN VALKENBURG. (Communicated by Prof. L. BOLK).

The woman, whose brain was examined, represented a case of not very grave idiocy, without important motor, sensible and sensory troubles. Causa mortis: tuberculosis pulmonum, at 38 years [1]).

Weight of the whole brain 390 grams.

The surface of the hemispheres shows a striking resemblance to the condition presented by the human foetus specially in the frontal part; in the parieto-occipital region the development seems more anthropoid.

From a confrontation of several foetal brains with anthropoid and normal human brains the following statements result.

The few furrows, (almost exclusively primary ones are present show for the greater part the simple condition as presented in foetus: they limit broad circumvolutions which give to the brain a macrogyric character, specially in the frontal and temporal lobi.

The parietal-occipital region shows the greatest lack of development, as is proved by the greater abnormalities in gyri and sulci.

Specially in the parieto-occipital "plis de passage" the abnormalities show a tendency in the direction of the anthropoid type. It is easy to see the character of the simian split in the system of the neighbouring furrows.

A simian split is present, when the parieto-occipital plis de passage are entirely or partly operculated by the occipital cortex. Sensu strictiori one cannot speak of a fissure in such a case, (as neither can be done with the sulci operculares insulae). Already this fact is sufficient to prove, that in principle it is wrong to accept in the human brain a simian split in those cases where the fiss. par. occ. is continuous with a transversal fissure on the par. occ. convexity as is generally done. (A similar condition is found in our case on the right side of the brain).

A continuation of the posterior wall of fissure *a* in a frontal direction over the anterior furrow of the delta-shaped complex would give rise to the formation of an operculum covering the entrance to the simian split.

In the condition as it is now the fissure *a* can be only considered as the caudal border of a virtual simian split. —

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[1]) From the cases described in detail, only one patient reached a higher age (49 years): case from HILTY, (v. MONAKOW's laboratory); the great majority were young children.

I draw attention to the fact, that the *sulcus calloso-marginalis* on the mesial side of the hemisphere does not show any fragmentation; and that there is present a broad pli de passage between *cuneus* and *praecuneus*. The confluency of *calcarina* and *parieto-occipitalis*, which is a general feature of the human brain, and is seen only very rarely in the brain of the highest anthropoids, is clearly shown in our cause on both hemispheres.

On an average the homology of the furrows and circumvolutions present in this idiot with those of the normal surface of the human brain, does not present great difficulties. The greatest of them is the interpretation of the poorly developed occipital region. In this part we see at first a continuation of t_2 from the temporal lobe; further the lateral ending of the fissura calcarina coming from the mesial surface. Next to these sulci a transverse furrow is observed in the left occipital lobe, possibly corresponding with the furrow x in the nomenclature of ZIEHEN and KÜKENTHAL; its occipital ending might have been pushed laterad by the strong outgrowth of the calcarina on the convexity.

For further details I merely refer to the explanation of the figures. Sometimes a more foetal, sometimes a more anthropoid type of sulci is seen, which mostly have a more casuistic than general importance.

The aspect of the *insulae Reilii* and the neighbouring parts is of greater interest. On both hemispheres the *insula* has remained uncovered for a large part. As has been pointed out by BOLK [1]) the human insula and its surroundings can be understood only by the study of the anthropoid apes, where the *sulcus opercularis* and the *fissura fronto-orbitalis* very often show a confluence.

Only by this confluence the operculisation of the insula becomes possible. This phylogenesis of the covering of the insula does not seem to be repeated ontogenetically, not even in its principle features. On the contrary in man no trace is found of the fissura fronto-orbitalis, by the inclusion of which in the insular space the homology of its frontal bifurcation with the two anterior branches of the fissura Sylvii would be explicable. A six-months human embryo showing on the convexity of its brain only the fissura Rolando, already exhibits in the anterior dorsal corner of the fossa Sylvii that small horizontal continuation, which, split up sagittally, afterwards represents both *rami anteriores*; whereas the ventrofrontal surface of the uncovered part of the insula still shows a direct continuation with the frontal lobe. The sagittal splitting of the above mentioned frontal continuation

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[1]) Beiträge zur Affenanatomie II (Orang Utan) in Petrus Camper I 1901 and VII (Gorilla) in Zeitschr. f. Morphol. ü Anthropol. Bd. XIII 1909.

of the fossa gives rise to the formation of the operculum triangulare in most human brains. On the other hand the separation of the insula surface from the frontal cortex by the sulcus opercularis in human embryos is already complete long before the operculisation is achieved. From this it results that there are *two differences* between the development of this region in phylogenetic and ontogenetic sense.

1. The confluence of the sulcus opercul. with the sulc. fronto-orb. which phylogenetically is the condition of the total operculisation of the frontal cortex and of the formation of the rr. ant. foss. Sylvii, apparently is not required in embryologic development, the sulc. fr. orb. wanting in human foetus.

2. An entire separation of the insula from the adnexa is not necessarily followed by total operculisation. The latter is demonstrated on the left hemisphere of our case where the fronto-orbital cortex is clearly separated from the insula surface by a rather deep furrow.

Looking for an homology with the rr. anteriores one might be inclined to consider the furrows α and β as such. Topographically the fact that they seem to limit the operc. triangulare might confirm this supposition.

If, however, we consider the structure of the right hemisphere, it appears that this operculum clearly shows a pli de passage between insula and frontal cortex (figure 3 ↙). Considering the way by which the rr. ant. and the operc. triang. are formed in the foetus we may exclude the possibility that a part of a cortex that has already been transformed into operculum, should become again immediately continuous with the insula simply by a cortex-bridge.

It seems to be more probable that the caudal expansion of the frontal cortex in this abnormally developed brain was especially in the right hemisphere so irregular, that instead of one, two separated sulci operculares were formed, which do not meet in the later development. On the right side the opercular sulcus, necessary for the formation of an operculum orbitale does not occur. Both insulae show the particularity that they cohere directly with the region behind the fiss. Rolando by a large circumvolution ; only the frontal part of the operculum Rolandicum is partly formed.

The question is, *to which of the surrounding surfaces, that usually take part in covering the insula, the unsufficient operculisation is due.* Where the latter has not developed at all, the answer can easily be given.

Certainly the retrorolandic cortex has failed in growth. This however does not explain the uncovered condition of the ventral part of the insula.

In order to point out the failing cortex-part it is necessary to have a punctum fixum, which has a constant position in the topographic constellation.

It seems to be allowable to regard as such a point the frontal border of the *foramen Monroi*. This choice is certainly justified by embryologic considerations (constant position of the lamina terminalis) [1]).

The projection of this point on the lateral wall of the hemisphere appears to lie nearly in the fiss. praecentralis, about 2 centimetres dorsally to the ventral border of the operc. frontale.

In our case the latter distance is much smaller, the point of projection falls only one millimeter in front of the junction of the horizontal and vertical sulci operculares. In regard to the ventral-orbital border of the operculum its position is not the same on the left and right hemisphere. On the left the normal condition is found, but on the right the ventral-orbital border of the operculum appears to be shifted in a frontal direction. Correcting in this way the insufficiency in the opercularisation above mentioned we see that only the ventral insula-part remains uncovered. If my supposition that this defect is not due to the insufficient expansion of the frontal cortex, is correct, only a subnormal development of the temporal lobe in a frontal direction can be considered to be the cause. Controlling this presumption by the topical relation of the temporal pole to the above mentioned projection-point we find the horizontal distance to be about 1 cm. less than in normal brains. — Bolk l.c. calls attention to the phylogenetic increase forwards of the temporal lobe; the fissura Rolando in its ventral continuation is used by him as a topic criterium. In our case I thought it better not to use this criterium. (N.B. the asymmetry of both fiss. centrales!); in another way however, a similar result was obtained.

If the extension of the temporal lobe is corrected in the drawings, not only an operculisation of the insula is obtained but also a more normal direction of the fissura Sylvii instead of the almost vertical furrow, of which only the posterior part represents the fissura Sylvii, whereas its frontal part is only a fold between the island and the temporal cortex.

The particular aspect of the surface of the brain is the expression of the *internal structure of the cortex*. The latter again is determined by the general laws of development in regard to eventual abnor-

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1) The limits of the corp. callosum — although used in the division of normal surfaces (v. Monakow) – cannot here be made use of, because, proportionally to the surface of the brain (the parts of which should be topographically determined) it varies in size.

malities in origin and ontogenetic evolution, as also by the quantity and the character of the connections between the cortical regions themselves and between these and subcortical centra. As to the latter the investigation of the fibre-tracts will give light.

Concerning the structure of the cortex some points can be stated even now, based on the examination of 14 cortex-pieces, taken from different regions and stained by NISSL's method.

No local pathological changes — diseases of vascular system, traces of inflammation etc. — could be found.

The resemblance with foetal structure is obvious: the layer of *multiform cells* (VI BRODMANN) extends too deep in the underlying centrum ovale. In this region ganglioncells are found, of which some have not yet attained to their full development; others have required the pyramidal form; apparently, on their way from the ependymary matrix to the perifery, they have not reached their destination (*heterotopy* in its simplest form).

In the *frontal cortex*, specially in the gyr. centr. ant. the *inner granular layer* (IV) is clearly visible in contrast to normal brains; evidently a remnant of the embryological conditions.

On the right hemisphere, the *area gigantopyramidalis* occupies a more frontal position to the fiss. centralis than in normal brain: the gigantic pyramids only begin on the top of the frontal border of this fissure, whereas on the left hemisphere they are already present on the bottom of the sulcus, and even penetrate into the region of gyr. centr. posterior.

Behind the fiss. centralis the changes are more obvious, at least in so far, that in more places than in the frontal lobe, there is an under-development of the *supragranular pyramids* [1]), whereas the infragranular ones show a tolerably regular condition (gyr. centr. post., T_1 and T_2 caudal parts). This abnormality is most striking on the bottom of the sulci where already *de norma* these, just as the deeper layers, are more poorly developed, compared with the enlarged lam. zonalis and granularis externa.

Those regions, having normally a very characteristic structure as the acoustic and visual cortex, show in this idiot the same general features. It should be mentioned that the *area striata* can be easily limited on the tops of the dorsal, resp. the central border of the *fissura calcarina*, where it touches the so-called visuo-psychic zone,

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1) In regard to the phylogenesis (see especially MOTT: Evolution of the visual cortex in Mammalia, Arch. of Neurol. Vol. III, and WATSON: The mammalian cerebral cortex etc., ibidem), this condition may be considered as a partial explanation of idiocy, apart from the general marked reduction in size of the brain.

recognisable by the loss of the superior half of the inner granular layer, which is characteristic of the calcarina, and which gives place to a row of large pyramidal cells. The line of VICQ D'AZIR at the same time is wanting.

So we find a human type in the structure of the occipital lobe the surface of which reminds us much more of the anthropoid brain.

EXPLANATION OF THE FIGURES.

The drawings have been made natural size by MARTIN's dioptrograph, and reduced to $^2/_3$.

Fig. 1. Dorsal aspect of the hemispheres.

c = *fiss. centralis*. On the right side nearly rectilinear, on the left side showing two frontal convexities

Between the third and fourth quarter a side-branch proceeds on both sides in a frontal direction. On the left side there seem to be two in the horizontal projection. The lateral limit however of the small square, which has been drawn as a furrow, is really a shallow fold in the lateral wall of the said side branch of which latter the mesial wall is made discontinuous by the confluence with the fissure parallel to *c* which has moreover "pulled mediad" the mesial wall of the same side branch. The widening is accompanied by a dislocation of the sulcus-bottom laterad, which produces the obliquity of the lateral wall, on which latter appears the mentioned shallow fold. — The whole makes a somewhat pathological impression. The part of it, that is parallel to *c* seems to be the only remnant of the *fissura praecentralis*.

pr = *fiss. praecentralis* (on the right side). The part that corresponds to the *fiss. praec. inferior* is continuous with the *sulc. opercularis* (fig. 3).

f_1 = *fiss. frontalis superior*, broken on both sides.

f_2 = *fiss. frontalis medius*.

rc = *fiss. retrocentralis*. On the left side it shows a posterior convexity apparently caused by the mesial part being bent in a frontal direction, in consequence of the encroachment on the convexity of the

cm = *fiss. calloso-marginalis*, which on the right side appears on the convexity in a more caudal region.

ip = *fiss. interparietalis*, on the left side it shows in contrast with the right a continuation with *r.c.*; on the left side it is broken. It is continuous with

a = *simian fissure* (see text).

p-o l = *fiss. parieto-occipitalis lateralis* appearing on the convexity. On the right side it is continuous with *i-p*.

t_1 = *fiss. temporalis superior*.

Figure 2. Lateral wall of the left hemisphere.

Figure 3. Lateral wall of the right hemisphere.

S = *fiss. Sylvii* (see text).

The furrows of the insula are dotted.

α, β = *sulc. opercularis* (see text).

↙ = direct continuation from cortex to insula (see text).

t_2 = *fiss. temporalis media*, only visible on the right hemisphere.

Figure 4. Mesial wall of the left hemisphere.
Figure 5. Mesial wall of the right hemisphere.
c-m = *fiss. calloso-marginalis*. Broken neither on the left side nor on the right.
r = *fiss. rostralis.*
rh = *fiss. rhinalisposterior.*
p-o = *fiss. parieto-occipitalis*. On the left side a broad gyrus cuneo-praecunealis is observed, which on the right side is only indicated as the beginning of a deep pli de passage about on the middle of *p-o*.
ca = *fiss. calcarina*, confluent with *p-o*.
cc = *corp. callosum.*
Not indicated are: *foramen Monroi*; immediately in front of this: *commissura anterior*. On the base the *optic chiasma* and the *mammilary bodies*. On the left side behind the thalamus the *pulvinar*.
Figure 6. Ventral wall of the left hemisphere
Figure 7. Ventral wall of the right hemisphere.
co = *fiss. collateralis* (= occipitotemporalis).
t_3 = *fiss. temporalis inferior*, which extends in anterior direction in front of the posterior part of the large fiss. rhinal. posterior (apes!).
f_3 = *fiss. frontalis inferior* (incisure and H. [Broca]).

Mathematics. — "*Investigation of the functions which can be built up by means of infinitesimal iteration. Contribution to the solution of the functional equation of* Abel." By M. J. van Uven. (Communicated by Prof. W. Kapteyn).

If we regard by the side of $y_1 = \varphi_1(x) = \varphi(x)$ also $y_2 = \varphi\{\varphi(x)\} = \varphi_2(x)$, $y_3 = \varphi\{\varphi_2 x)\} = \varphi[\varphi\{\varphi(x)\}] = \varphi_3(x)$ etc. . . . $y_n = \varphi_n(x)$ . . ., we can ask ourselves whether this "iteration" of the function $\varphi(x)$ leads to a definite final value y_∞, when it is continued to infinity and how this final form behaves from the point of view of the theory of functions. In this sense the problem of iteration has been taken by numbers of mathematicians, among others by E. Schroeder [1]), Koenigs [2]), Farkas [3]), Isenkrahe [4]), Bourlet [5]), Léméray [6]), Carlsson [7]). Their considerations lead more than once to the construction of new algorithms to approximate roots of algebraical and transcendental equations. Others on the other hand have put the question: which function

[1]) Math. Ann. Bd. II (1870), p. 317.
[2]) Bulletin des sc. math. t. VII (1883), p. 340; Ann. scientif. de l'Ecole norm. sup. (3) t. I (1884), suppl. p. 3; t. II (1885), p. 385.
[3]) Journal de mathém. (3) t. X (1884), p. 101.
[4]) Math. Ann. Bd. XXXI (1888), p. 309. Das Verfahren der Funktionswiederholung, Teubner, 1897.
[5]) Ann. d. l. Fac. des sc. de Toulouse t. XII (1898), C.
[6]) Among others Bulletin d. l. Soc. Math. t. XXVI (1898), p. 10.
[7]) Om itererade funktioner. Diss. Uppsala, 1907.

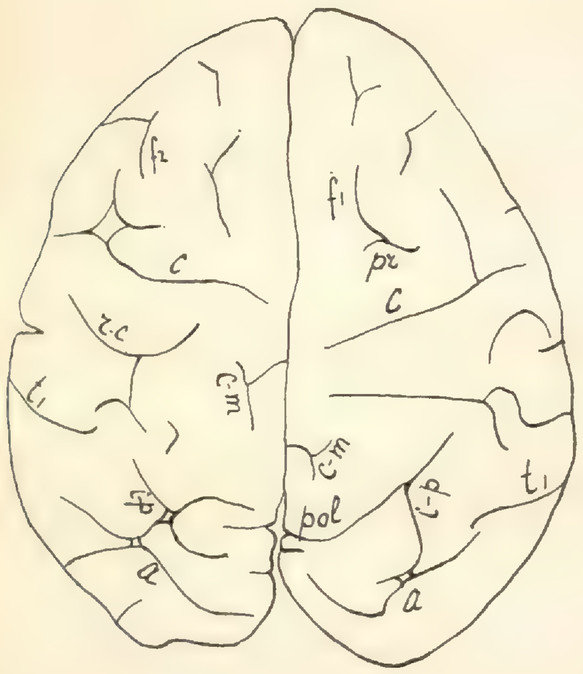

Fig. 1.

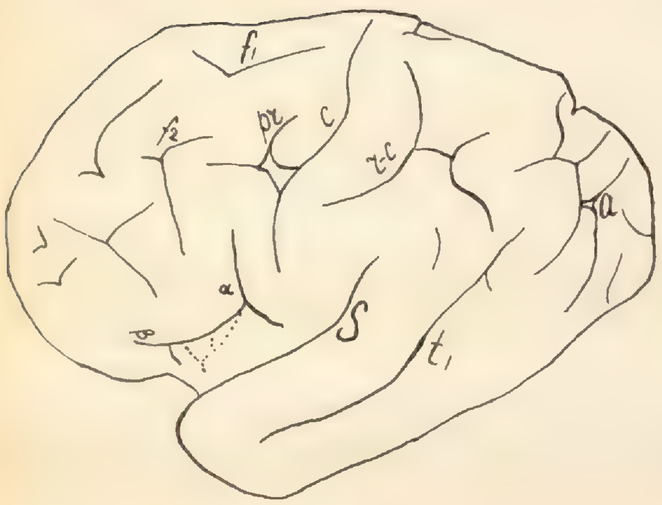

Fig. 2.

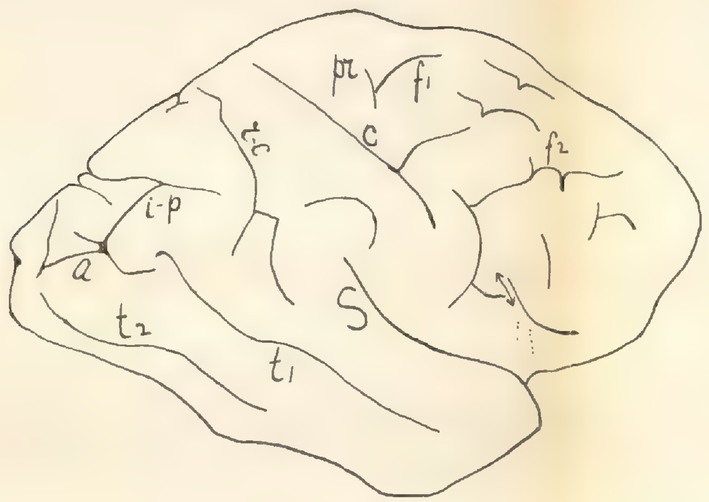

Fig. 3.

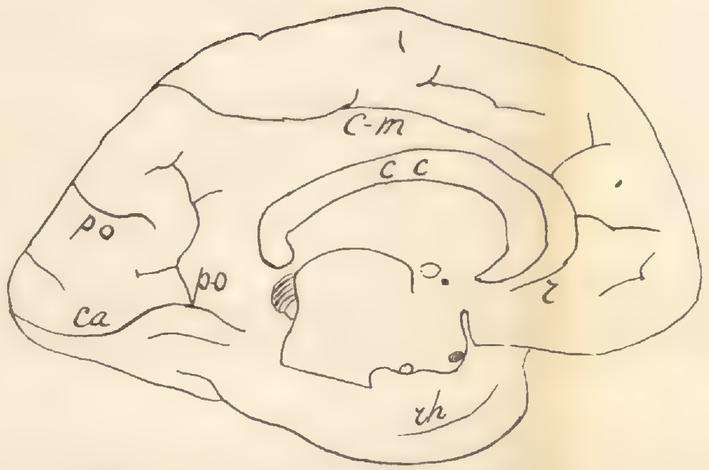

Fig. 4.

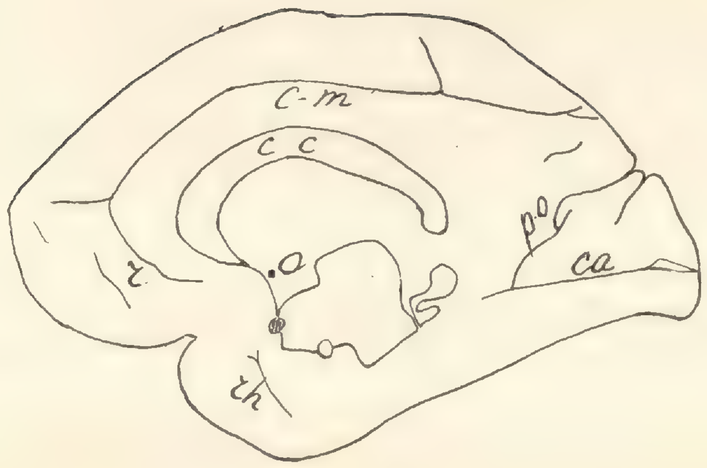

Fig. 5.

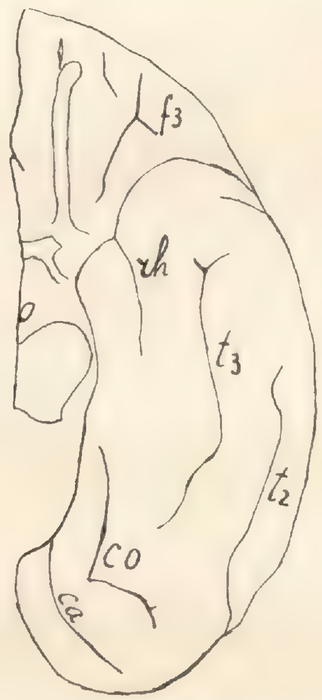

Fig. 6.

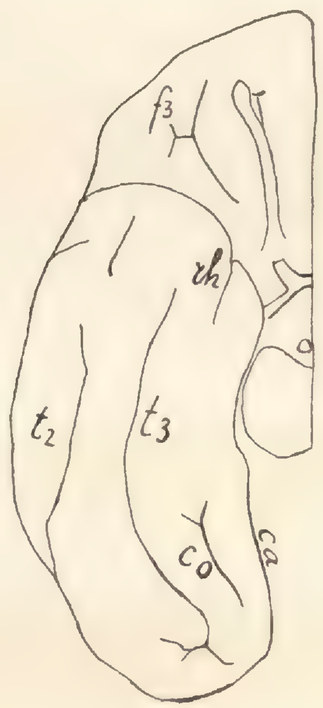

Fig. 7.

$\psi(x)$ passes after an iteration $n-1$ times repeated into a given function $\varphi(x)$, so that $\psi_n(x) = \varphi(x)$ is satisfied. With the answer to this question among others E. SCHROEDER[1]), FORMENTI[2]), LEAU[3]), SPIESS[4]) have occupied themselves.

We too will take the problem of iteration from this side and we will endeavour to find an expression for the $(n-1)^{st}$ iteration $\varphi_n(x)$ of $\varphi(x)$ which retains its importance for *all real* values of the iterationindex n. When we have found that expression we have but to put $n = \frac{1}{m}$ to obtain the function $\psi = \varphi_{\frac{1}{m}}$, furnishing φ after an iteration $n-1$ times repeated so that

$$\psi_m(x) = \varphi_{\frac{1}{m}.m}(x) = \varphi_1(x) = \varphi(x).$$

By increasing n with infinitesimal amounts we can as it were build up the function φ by *infinitesimal iteration.*

It is necessary to just distinguish here between two kinds of infinitesimal iteration. If namely we consider (see SPIESS)[4]) the *operation* $x + \delta x$, where $\delta = \underset{m=\infty}{Lim} \frac{p}{m}$, then by iterating this *operation* $(m-1)$ times we obtain $Lim\ (1+\delta)^m x = x \,.\, e^p$. We on the contrary wish to occupy ourselves with the case in which not an operation but a *function* proper is iterated.

It is evidently the point to express $\varphi_n(x)$ in the form of a continuous function of x and n. However we can put the problem also like this: which form must function $f(x)$ have to be increased with *unity* by the substitution $y = \rho(x)$ instead of x, i. o. w.: for which function $f(x)$ holds for a given $y = \varphi(x)$

$$f(y) = f\{\varphi(x)\} = f(x) + 1, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (A)$$

therefore also

$$f(\varphi_n) = f(x) + n\ ?$$

If the function $f(x)$ is determined, then φ_n follows out of

$$\varphi_n(x) = f_{-1}\{f(x) + n\}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (a)$$

in which f_{-1} represents the *inverse* function of f. We have then reached our aim: we have written φ_n as function of x and n.

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[1]) Math. Ann. Bd. III (1871), p. 296.

[2]) Rendiconti d. R. Istituto Lombardo (2) vol. VIII (1875), p. 276.

[3]) Ann. d. l. Fac. des sc. de Toulouse t. XI (1897), E.

[4]) Mitteil. der Naturforsch. Gesellsch. in Bern, 1901, p. 106.
See also the article of PINCHERLE in the Encycl. der Math. Wiss. Bd. II 1, Heft 6, p. 791.

It is our task therefore to solve the functional equation (A). This equation is called the *functional equation of* ABEL [1]). It is to be regarded as the logarithmic form of the equation

$$F\{\varphi(x)\} = \alpha F(x),$$

which bears the name of the *equation of* SCHROEDER [2]).

Whilst LEAU regards the solution of the equation of ABEL (SCHROEDER) from the point of view of the theory of functions and FORMENTI and SPIESS confine themselves to a few special cases, SCHROEDER deduces some groups of functions $\varphi(x)$ for which the function $f(x)$ is easy to determine. It is now our intention to give to the results of SCHROEDER a more general form and to furnish in this way a contribution to the solution of the two functional equations mentioned.

A first group of functions $y = \varphi(x)$ for which φ_n is easy to determine is obtained *by choosing for* $f(x)$ *a definite function.*

Example I:

$$f(x) = x^p,\ y^p = x^p + 1,\ y = \varphi(x) = \sqrt[p]{x^p + 1},\ \varphi_n(x) = \sqrt[p]{x^p + n}.$$

Example II;

$$f(x) = \sin x;\ \sin y = \sin x + 1,\ y = \varphi(x) = \sin^{-1}(\sin x + 1),\ \varphi_n(x) = \sin^{-1}(\sin x + n).$$

It is as easy, however, to find the function $f(x)$ and with it the expression for φ_n *when in the equation*

$$g(y) = g(x) + \beta \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (B)$$

we choose for $g(x)$ *a definite function.* For, in this case $f(x) = \frac{g(x)}{\beta}$ and

$$\varphi_n(x) = g_{-1}\{g(x) + n\beta\} \quad . \quad . \quad . \quad . \quad . \quad . \quad (b)$$

Example III:

$$g(x) = \log x,\ \log y = \log x + \beta = \log x + \log \gamma, \qquad (\gamma = e^{\beta})$$

$$y = \varphi(x) = \gamma x \quad ; \quad \varphi_n(x) = \gamma^n x.$$

If we were able to write every function $y = \varphi(x)$ in the form (B), the problem of the solution of the functional equation of ABEL would be very simple. It is clear that we stand a greater chance of bringing a function $y = \varphi(x)$ in a certain standard form when this form contains more arbitrary constants. *It is our aim therefore to increase the number of constants in the standard form.*

Though a *given* function resists a transformation in the standard form found by us, no real difficulties adhere to the problem, "to

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[1]) Oeuvres complètes II, p. 36.

[2]) See note 1 p. 208.

construct as many functions $\varphi_n(x)$ as one likes", which in fact may be called solved in (a) and (b).

§ 2. We shall now pass on to the investigation of a new standard form. We first imagine the function $y = \varphi(x)$ to be represented by the two parameterequations

$$x = \xi(t) \quad , \quad y = \eta(t) \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

As the function $f(x)$ is so to say the connecting link between the function φ and its iteral φ_n, a relation, e.g. a differential equation, which is satisfied by all φ_n, so also by $\varphi_0(x) = x$, may be written as an equation, in which the function f is the independent variable, thus as a relation of the form

$$\Phi\left(f \ , \ \varphi_n \ , \ \frac{d\varphi_n}{df} \ , \ \frac{d^2\varphi_n}{df^2} \ , \ \ldots\right) = 0.$$

If here we exchange the independent variable with the dependent variable then we must be able to arrive at an equation

$$F\left\{\varphi_n \ . \ f(\varphi_n) \ , \ \frac{df}{d\varphi_n} \ , \ \frac{d^2f}{d\varphi_n^2} \ , \ \ldots .\right\} = c,$$

retaining the same, so a *constant* value for all φ_n, therefore also for $\varphi_0(x) = x$; i. o. w. out of (1) and (A) a relation must be deduced of the form

$$F\{x, f(x), f'(x), f''(x), \ldots\} = F\{y, f(y), f'(y), f''(y), \ldots\} =$$
$$= \ldots = F\{y_n, f(y_n), f'(y_n), f''(y_n) \ldots\} = c. \quad . \quad . \quad . \quad (2)$$

In F no forms will be allowed to appear, in which a strange variable is present, therefore not $\frac{dy}{dx}$, $\frac{dx}{dt}$, $\frac{dy}{dt}$ etc.

By differentiating (A) according to t we find

$$\varrho = f'(y)\, y' = f'(x)\, x' . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

A first (trivial) solution we get by putting: $y' = x'$, or $y = x + \beta$; $f'(y) = f'(x) = c$, $f(x) = cx + c'$, where (A) is then satisfied, when we put $c = \frac{1}{\beta}$, $c' = 0$, therefore $f(x) = \frac{x}{\beta}$; $\varphi_n = x + n\beta$.

Differentiation of (3) produces

$$\varrho' = f''(y)\, y'^2 + f'(y)\, y'' = f''(x)\, x'^2 + f'(x)\, x'',$$

or, on account of (3)

$$\frac{\varrho'}{\varrho} = \varrho \frac{f''(y)}{f'^2(y)} + \frac{y''}{y'} = \varrho \frac{f''(x)}{f'^2(x)} + \frac{x''}{x'},$$

or

$$\varrho \left\{ \frac{f''(y)}{f'^2(y)} - \frac{f''(x)}{f'^2(x)} \right\} = -\left(\frac{y''}{y'} - \frac{x''}{x'}\right) . \quad . \quad . \quad . \quad . \quad (4)$$

To arrive at an equation of the form (2) the second member of (4) must be reducible by means of an equation obtained by differentiation of (1) to a form $\Psi(y, y', y'', \ldots) - \Psi(x, x', x'', \ldots) = \Psi_y - \Psi_x$, which in its turn must allow of being brought to the form $\varrho\,[\Phi\{y, f(y), f'(y), f''(y), \ldots\} - \Phi\{x, f(x), f'(x), f''(x)\ldots\}] = \varrho\,(\Phi_y - \Phi_x) = y' f'(y)\,\Phi_y - x' f'(x)\,\Phi_x$, so that $\Psi_y - \Psi_x = y' f'(y)\,\Phi_y - x' f'(x)\,\Phi_x$. From this ensues that $f'(y)\,\Phi_y$ may contain y, but not $f(y)$, $f'(y)$,... or i. o. w. that $f'(y)\,\Phi_y = \psi(y)$ and then likewise $f'(x)\,\Phi_x = \psi(x)$ must hold; therefore Φ_y must be of the form $\psi(y) : f'(y)$ and Φ_x of the form $\psi(x) : f'(x)$. Hence the transformation of the second member of (4) looks as follows

$$-\left(\frac{y''}{y'} - \frac{x''}{x'}\right) = \psi(y) \cdot y' - \psi(x) \cdot x' = \varrho\left\{\frac{\psi(y)}{f'(y)} - \frac{\psi(x)}{f'(x)}\right\}.$$

If therefore the given function $y = \varphi(x)$, represented by (1), satisfies the differential equation of the second order

$$\frac{y''}{y'} - \frac{x''}{x'} + \psi(y) \cdot y' - \psi(x) \cdot x' = 0, \quad \ldots\ldots \quad (5)$$

then $f(y)$ (resp. $f(x)$) is determined out of

$$\frac{f''(y)}{f'^2(y)} - \frac{f''(x)}{f'^2(x)} = \frac{\psi(y)}{f'(y)} - \frac{\psi(x)}{f'(x)},$$

i. e. out of

$$\frac{f''(x)}{f'^2(x)} - \frac{\psi(x)}{f'(x)} = c, \quad \ldots\ldots\ldots \quad (6)$$

and we are able to build up the function φ by infinitesimal iteration.

The equation (5) passes when we put $t = x$ into

$$\frac{y''}{y'} + \psi(y) \cdot y' - \psi(x) = 0, \quad \ldots\ldots\ldots \quad (7)$$

from which ensues by integration

$$g(y) = \alpha\, g(x) + \beta, \quad \ldots\ldots\ldots \quad (C)$$

where $g(x)$ is put equal to $\int e^{\int \psi(x)\,dx}\,dx$.

With this we have obtained a new standard form, one *with two arbitrary constants*. By putting either $\alpha = 1$, or $\beta = 0$ we get out of this the two standard forms of SCHROEDER (l. c. p. 303).

We have now but to bring a given function $y = \varphi(x)$ into the form (C) or by differentiation into one of the forms (7) or

$$y' = \alpha\,\frac{g'(x)}{g'(y)} = \alpha\,\frac{h(x)}{h(y)} = \alpha\,\frac{H(y)}{H(x)} \quad \ldots\ldots \quad (8)$$

By integrating (6) we find in connection with (A),

$$f(x) = \frac{log\ a}{log\ \alpha} + \frac{log\left\{g(x) + \frac{\beta}{\alpha-1}\right\}}{log\ \alpha},$$

or if we put $a = 1$

$$f(x) = \frac{log\left\{g(x) + \frac{\beta}{\alpha-1}\right\}}{log\ \alpha}. \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

Out of (a) then follows for $\varphi_n(x)$

$$\varphi_n(x) = g_{-1}\left\{\alpha^n g(x) + \frac{\alpha^n-1}{\alpha-1}\beta\right\}. \quad . \quad . \quad . \quad . \quad . \quad (c)$$

By putting $g(x) = \zeta$ and therefore $x = g_{-1}(\zeta) = G(\zeta)$ we can reduce the standard form (C) to the form:

$$x = G(\zeta) \quad , \quad y = G(a\zeta + \beta), \quad . \quad . \quad . \quad . \quad . \quad (C')$$

or written more symmetrically,

$$x = G(a_1\zeta + a_2) \quad , \quad y = G(b_1\zeta + b_2). \quad . \quad . \quad . \quad . \quad (C'')$$

So if we succeed in giving to $y = \varphi(x)$ this parameter representation, the function can be built up by infinitesimal iteration.

The expression for $f(x)$ loses its signification when $log\ \alpha = 0$, so when $\alpha = 1$. We are then however in the case (B), where $f(x) = \frac{g(x)}{\beta}$ which form follows likewise out of (7) when there c is put equal to $log\ \alpha = 0$. The form for $\varphi_n(x)$ we can also obtain by putting α equal to 1 in (c). Then holds $\underset{\alpha=1}{Lim}\ \frac{\alpha^n-1}{\alpha-1} = n$, so that $\varphi_n(x)$ assumes the form (b).

§ 3. Before passing on to giving some examples we shall endeavour to deduce along the same way differential equations of a higher order, to arrive at more arbitrary constants in the standard form.

By differentiation of (4) we arrive at

$$\varrho'\left\{\frac{f''(y)}{f'^2(y)} - \frac{f''(x)}{f'^2(x)}\right\} + \varrho\left[\left\{\frac{f'''(y)}{f'^2(y)}y' - 2\frac{f''^2(y)}{f'^3(y)}y'\right\} - \left\{\frac{f'''(x)}{f'^2(x)}x' - 2\frac{f''^2(x)}{f'^3(x)}x'\right\}\right] =$$

$$= -\left(\frac{y'''}{y'} - \frac{x'''}{x'}\right) + \left(\frac{y''^2}{y'^2} - \frac{x''^2}{x'^2}\right),$$

or if we put:

$$\frac{f''(x)}{f'^2(x)} = X_2, \frac{f''(y)}{f'^2(y)} = Y_2, \frac{f'''(x)}{f'^3(x)} = X_3, \text{ ect. } \frac{y''}{y'} = \eta_2, \frac{x''}{x'} = \xi_2, \frac{y'''}{y'} = \eta_3, \text{ etc.,} \quad (10)$$

$$\varrho'(Y_2 - X_2) + \varrho^2\{(Y_3 - 2Y_2^2) - (X_3 - 2X_2^2)\} = -\{(\eta_3 - \eta_2^2) - (\xi_3 - \xi_2^2)\}. \quad (11)$$

Now we have on account of (3) and (10)

$$\varrho' = \varrho^2 Y_2 + \varrho\eta_2 = \varrho^2 X_2 + \varrho\xi_2 = \tfrac{1}{2}\varrho^2 (Y_2 + X_2) + \tfrac{1}{2}\varrho (\eta_2 + \xi_2),$$

$$(Y_2 - X_2) = -\frac{1}{\varrho}(\eta_2 - \xi_2),$$

so that we can write:

$$\varrho'(Y_2 - X_2) = \tfrac{1}{2}\varrho^2 (Y_2 + X_2)(Y_2 - X_2) + \tfrac{1}{2}\varrho(\eta_2 + \xi_2) \cdot -\frac{1}{\varrho}(\eta_2 - \xi_2) =$$

$$= \tfrac{1}{2}\varrho^2 (Y_2^2 - X_2^2) - \tfrac{1}{2}(\eta_2^2 - \xi_2^2),$$

by which (11) passes into

$$\varrho^2 \left\{\left(Y_3 - \frac{3}{2} Y_2^2\right) - \left(X_3 - \frac{3}{2} X_2^2\right)\right\} = -\left\{\left(\eta_3 - \frac{3}{2}\eta_2^2\right) - \left(\xi_3 - \frac{3}{2}\eta_2^2\right)\right\}. \quad (12)$$

The second member of (12) must now be brought by means of the differential equation of $y = \varphi(x)$ into the form

$$\varrho^2 [\Phi \{y, f'(y), \ldots\} - \Phi \{x, f'(x), \ldots\}];$$

from this can easily be found that $\Phi(y)$ must be of the form $\omega(y) : f'^2(y)$; the transformation of the second member of (12) takes place as follows

$$-\left\{\left(\frac{y'''}{y'} - \frac{3}{2}\frac{y''^2}{y'^2}\right) - \left(\frac{x'''}{x'} - \frac{3}{2}\frac{x''^2}{x'^2}\right)\right\} = \omega(y)y'^2 - \omega(x)x'^2 = \varrho^2 \left\{\frac{\omega(y)}{f'^2(y)} - \frac{\omega(x)}{f'^2(x)}\right\}.$$

If now the given function $y = \varphi(x)$ satisfies the differential equation of the third order

$$\left(\frac{y'''}{y'} - \frac{3}{2}\frac{y''^2}{y'^2}\right) - \left(\frac{x'''}{x'} - \frac{3}{2}\frac{x''^2}{x'^2}\right) + \omega(y)y'^2 - \omega(x)x'^2 = 0, \quad . \quad (13)$$

then $f(x)$ resp. $f(y)$ is found out of

$$\left\{\frac{f'''(y)}{f'^3(y)} - \frac{3}{2}\frac{f''^2(y)}{f'^4(y)}\right\} - \left\{\frac{f'''(x)}{f'^3(x)} - \frac{3}{2}\frac{f''^2(x)}{f'^4(x)}\right\} = \frac{\omega(y)}{f'^2(y)} - \frac{\omega(x)}{f'^2(x)},$$

so out of

$$\frac{f'''(x)}{f'^3(x)} - \frac{3}{2}\frac{f''^2(y)}{f'^4(x)} - \frac{\omega(x)}{f'^2(x)} = c, \quad . \quad . \quad . \quad . \quad . \quad . \quad (14)$$

and then here too we are able to build up $\varphi(x)$ by means of infinitesimal iteration.

By putting t equal to x (13) passes into

$$\frac{y'''}{y'} - \frac{3}{2}\frac{y''^2}{y'^2} + \omega(y)y'^2 - \omega(x) = 0. \quad . \quad . \quad . \quad . \quad . \quad (15)$$

The integration furnishes if we introduce for the integral of $\frac{F'''(x)}{F'(x)} - \frac{3}{2}\frac{F''^2(x)}{F'^2(x)} = \omega(x)$ the symbol $g(x)$

$$g(y) = \frac{\alpha g(x) + \beta}{\gamma g(x) + \delta}; \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (D)$$

thus a *new standard form with three independent arbitrary constants.*

As $\omega(x) = \frac{g'''(x)}{g'(x)} - \frac{3}{2}\frac{g''^2(x)}{g'^2(x)}$ we find by integration of (14)

$$f(x) = \frac{1}{\sqrt{2c}} log \frac{pg(x)+1}{qg(x)+1} + a = \frac{1}{\lambda} log \frac{pg(x)+1}{qg(x)+1} + a.$$

In order to satisfy (A) we dispose in such a way of the constants λ, p, q and a, that

$$f(y) = \frac{1}{\lambda} log \frac{pg(y)+1}{qg(y)+1} + a = \frac{1}{\lambda} log \frac{p\frac{\alpha g(x)+\beta}{\gamma g(x)+\delta}+1}{q\frac{\alpha g(x)+\beta}{\gamma g(x)+\delta}+1} + a = \frac{1}{\lambda} log \frac{pg(x)+1}{qg(x)+1} + a + 1.$$

To this end we must put

$$\left.\begin{aligned} p &= \frac{(\alpha-\delta) + \sqrt{(\alpha-\delta)^2+4\beta\gamma}}{2\beta} \quad , \quad q = \frac{(\alpha-\delta)-\sqrt{(\alpha-\delta)^2+4\beta\gamma}}{2\beta}, \\ \lambda &= log \frac{\{\alpha+\delta+\sqrt{(\alpha-\delta)^2+4\beta\gamma}\}^2}{4(\alpha\delta-\beta\gamma)} = log\, s, \end{aligned}\right\} \quad (16)$$

whilst we choose $a = 0$.

We then find for $f(x)$

$$f(x) = \frac{1}{\lambda} log \frac{pg(x)+1}{qg(x)+1}, \quad . \quad . \quad . \quad . \quad . \quad . \quad (17)$$

where p, q and λ are determined by (16).

Let us still put

$$\left.\begin{aligned} u &= \frac{\alpha+\delta}{2} \quad , \quad v = \frac{\sqrt{(\alpha-\delta)^2+4\beta\gamma}}{2} \left(\text{so that } s = \frac{u+v}{u-v} \right) \\ P(n) &= \frac{(u+v)^n-(u-v)^n}{2v} \quad , \quad Q(n) = (u+v)^n + (u-v)^n, \end{aligned}\right\} \quad . \quad (18)$$

we then find on account of (a) for $\varphi_n(x)$

$$\varphi_n(x) = g_{-1}\left[\frac{\{Q(n) + (\alpha-\delta)\,P(n)\}\,g(x) + 2\beta P(n)}{2\gamma P(n) g(x) + \{Q(n)-(\alpha-\delta)P(n)\}}\right] \quad . \quad . \quad (d)$$

By putting $g(x) = \zeta$ and therefore $x = g_{-1}(\zeta) = G(\zeta)$ we can make the standard form (D) to pass into

$$x = G(\zeta) \quad , \quad y = G\left(\frac{\alpha\zeta+\beta}{\gamma\zeta+\delta}\right), \quad . \quad . \quad . \quad . \quad . \quad (D')$$

or in a more symmetrical form,

$$x = G\left(\frac{a_1\zeta+a_2}{a_3\zeta+a_4}\right) \quad , \quad y = G\left(\frac{b_1\zeta+b_2}{b_3\zeta+b_4}\right) \quad . \quad . \quad . \quad . \quad (D'')$$

When thus we succeed in finding such a parameter ζ that $y = \varphi(x)$ can be brought to the form (D') or (D''), then for such a function φ the iterationproblem is solved.

The deduction of $f(x)$ chosen by us loses its validity as soon as we have

$$2v = \sqrt{(\alpha-\delta)^2 + 4\beta\gamma} = 0.$$

For then λ becomes $= 0$ and $p = q$. In this case however $c = 0$, so that the differential equation (14) now runs:

$$\frac{f'''(x)}{f'(x)} - \frac{3}{2}\frac{f''^2(x)}{f'^2(x)} = \frac{g'''(x)}{g'(x)} - \frac{3}{2}\frac{g''^2(x)}{g'^2(x)} \quad . \quad . \quad . \quad . \quad (19)$$

The integral of this is

$$f(x) = s'\frac{p'g(x)+1}{q'g(x)+1}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (20)$$

where, to satisfy (A) and (D) we must put

$$s' = 1 \quad , \quad p' = \frac{\alpha}{\beta} \quad , \quad q' = \frac{\alpha-\delta}{2\beta} \quad . \quad . \quad . \quad . \quad . \quad (21)$$

The expression for $\varphi_n(x)$ we find most easily by considering that $\underset{v=0}{P}(n) = nu^{n-1}$ and $\underset{v=0}{Q}(n) = 2u^n$. We thus arrive at

$$\varphi_n(x) = g_{-1}\left[\frac{\{(\alpha+\delta) + n(\alpha-\delta)\}g(x) + 2n\beta}{2n\gamma\, g(x) + \{(\alpha+\delta)-n(\alpha-\delta)\}}\right] \quad . \quad . \quad . \quad . \quad (d')$$

The case $v = 0$ forms the so-called *parabolic* case [1]) of the linear substitution, because the invariant points $g(y) = g(x)$ coincide.

Also of the standardform (D) we shall defer the applications until later.

§ 4. Beforehand we shall try to heighten the order of the differential equation by means of whose integration the standard form originated.

By differentiation of (12) we find with the aid of (3)

$$2\varrho\varrho'\{(Y_3 - \frac{3}{2}Y_2^2) - (X_3 - \frac{3}{2}X_2^2)\} + \varrho^3\{(Y_4 - 6Y_2Y_3 + 6Y_2^3) -$$

$$- (X_4 - 6X_2X_3 + 6X_2^3)\} = -\{(\eta_4 - 4\eta_2\eta_3 + 3\eta_2^3) -$$

$$- (\xi_4 - 4\xi_2\xi_3 + 3\xi_2^3)\}.$$

As farthieron holds: $\varrho' = \varrho^2 Y_2 + \varrho\eta_2 = \varrho^2 X_2 + \varrho\xi_2$, we can also write

$$\varrho^3\{(Y_4 - 4Y_2Y_3 + 3Y_2^3) - (X_4 - 4X_2X_3 + 3X_2^3)\} + 2\varrho^2\{(Y_3 - \frac{3}{2}Y_2^2)\eta_2 -$$

$$- (X_3 - \frac{3}{2}X_2^2)\xi_2\} = -\{(\eta_4 - 4\eta_2\eta_3 + 3\eta_2^3) - (\xi_4 - 4\xi_2\xi_3 + 3\xi_2^3)\}. \quad (22)$$

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[1]) Cf. KLEIN—FRICKE Vorles. ü. d. Theorie der Ell. Modulfunktionen, Teubner, 1890, p. 164.

Now it is impossible to bring the term with ϱ^2 into the form $\varrho^3 [\Phi\{y, f(y), f'(y), \ldots\} - \Phi\{x, f(x), f'(x), \ldots\}] + [\Psi(y, y', y'', \ldots) - \Psi(x, x', x'', \ldots)]$; in that way we can neither break up (22) into a differential equation for $y = \varphi(x)$ and one for $f(x)$.

The way chosen by us leads therefore no further than to the standard form (*D*) *with three arbitrary constants.*

An attempt to start from the equations (*C*) or (*D*) in order to treat them in the same manner as (*A*) and to find in that way new standard forms, offers no more chance of success.

If namely we differentiate

$$g(y) = \alpha g(x) + \beta$$

twice, then after elimination of the constants we find

$$\frac{g''(y)}{g'(y)} y' + \frac{y''}{y'} = \frac{g''(x)}{g'(x)} x' + \frac{x''}{x'} = \sigma \quad . \quad . \quad . \quad . \quad . \quad (23)$$

In this equation we must endeavour to separate by means of differentiation the expressions x', y', x'' etc. from the forms g', g'' etc. We first find

$$\frac{g'''(y)}{g'(y)} y'^2 - \frac{g''^2(y)}{g'^2(y)} y'^2 + \frac{g''(y)}{g'(y)} y'' + \frac{y'''}{y'} - \frac{y''^2}{y'^2} =$$

$$= \frac{g'''(x)}{g'(x)} x'^2 - \frac{g''^2(x)}{g'^2(x)} x'^2 + \frac{g''(x)}{g'(x)} x'' + \frac{x'''}{x'} - \frac{x''^2}{x'^2}.$$

As now however y' as well as y'' (resp. x' as well as x'') appear as factors of forms with $g'(y), g''(y), \ldots$ (resp. $g'(x), g''(x), \ldots$) and we have but *one* equation namely (23) at our disposal, we cannot separate the two expressions from each other, so that in this direction also further investigation will prove in vain.

We can therefore do nothing else but repeat the above final conclusion: with the method followed here we get no further than the standard form (*D*).

From all standard forms it is evident, that when $y = \varphi(x)$ can be built up by infinitesimal iteration, the same holds for $y = h_{-1}[\varphi\{h(x)\}]$, where $h(x)$ represents an *arbitrary* function of x. For $h_{-1}\{g_{-1}(x)\}$ is the inverse function of $g\{h(x)\}$. This result has even been found already by SCHROEDER (l.c. p. 301).

§ 5. Here may follow some examples.

Applications of (*C*) and (*c*):

Example IV. $g(x) = x$; $y = \varphi(x) = \alpha x + \beta$; $\varphi_n(x) = \alpha^n x + \frac{\alpha^n - 1}{\alpha - 1} \beta$.

Example V. $g(x) = log\, x\,;\; y = \varphi(x) = e^{\beta}x^{\alpha} = \gamma x^{\alpha}\,;\; \varphi_n(x) = \gamma^{\frac{\alpha^n-1}{\alpha-1}}\, x^{\alpha^n}$.

„ VI. $g(x) = log\,\dfrac{1-x}{1+x} = 2\, th^{-1}\, x\,;\; y = \varphi(x) = th\left(\alpha\, th^{-1}\, x + \dfrac{\beta}{2}\right)$;

$$\varphi_n(x) = th\left(\alpha^n\, th^{-1}\, x + \frac{\alpha^n-1}{\alpha-1}\cdot\frac{\beta}{2}\right)^{1)}.$$

„ VII. $g(x) = \dfrac{1}{x}\,;\; y = \varphi(x) = \dfrac{x}{\beta x+\alpha}\,;\; \varphi_n(x) = \dfrac{(\alpha-1)\,x}{(\alpha^n-1)\beta x+(\alpha-1)\alpha^n}$.

„ VIII. $g(x) = \dfrac{1}{x^2}\,;\; y = \varphi(x) = \dfrac{x}{\sqrt{\beta x^2+\alpha}}$;

$$\varphi_n(x) = \frac{x\sqrt{\alpha-1}}{\sqrt{(\alpha^n-1)\beta x^2 + (\alpha-1)\alpha^n}}.$$

„ IX. $g(x) = \sqrt{x}\,;\; y = \varphi(x) = \alpha^2 x + 2\alpha\beta\sqrt{x} + \beta^2\,;$

$$\varphi_n(x) = \alpha^{2n}x + 2\,\frac{\alpha^n-1}{\alpha-1}\,\alpha^n\beta\sqrt{x} + \left(\frac{\alpha^n-1}{\alpha-1}\right)^2\beta^2.$$

„ X. $g(x) = sin\, x\,;\; y = \varphi(x) = sin^{-1}\,(\alpha\, sin\, x + \beta)\,;$

$$\varphi_n(x) = sin^{-1}\left(\alpha^n\, sin\, x + \frac{\alpha^n-1}{\alpha-1}\,\beta\right).$$

Application of (D) and (d):

Example XI. $g(x) = x\,;\; y = \varphi(x) = \dfrac{\alpha x+\beta}{\gamma x+\delta}$;

$$\varphi_n(x) = \frac{\{Q(n) + (\alpha-\delta)P(n)\}x + 2\beta P(n)}{2\gamma P(n)x + \{Q(n) - (\alpha-\delta)P(n)\}}.$$

„ XII. $g(x) = x^2\,;\; y = \varphi(x) = \sqrt{\dfrac{\alpha x^2+\beta}{\gamma x^2+\delta}}$;

$$\varphi_n(x) = \sqrt{\frac{\{Q(n) + (\alpha-\delta)P(n)\}x^2 + 2\beta P(n)}{2\gamma P(n)x^2 + \{Q(n) - (\alpha-\delta)P(n)\}}}.$$

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1) SCHROEDER gives in a somewhat modified notation this example too, among others with $\alpha=2$, $\beta=0$, in consequence of which $y = th\,(2\, th^{-1}\, x) = \dfrac{2x}{1+x^2}\,;\; \varphi_n(x) = th\,(2^n\, th^{-1}\, x)$. (See Math. Ann. II p. 304)

Botany. — *"The influence of temperature on the respiration of the higher plants."* By J. KUYPER. (Communicated by Prof. F. WENT.)

In 1905 Miss MATTHAEI [1]) carried out some investigations on assimilation in the leaves of Prunus Laurocerasus. The figures thus obtained led BLACKMAN [2]) to some theoretical considerations entitled: "Optima and limiting factors."

According to these views there arises in the curve representing the variation of a physiological process with temperature an optimum, in consequence of a harmful influence at higher temperatures, which influence increases with the temperature. If the process were to follow the laws of a chemical reaction completely, then the rule of VAN 'T HOFF-ARRHENIUS would hold good for any temperature interval of 10°; the figures show, however, that in the case of assimilation this rule only applies between 0° and 20°—25°. If by this rule the theoretical values are calculated for higher temperatures, they prove to be much higher than those found experimentally. At the same time these values decrease, in proportion as the objects have been exposed for a longer time to the higher temperature. According to BLACKMAN the calculated value agrees with that found by exterpolation to zero time from the series of figures found by observation in successive equal time intervals at constant temperatures.

As a corollary of this view it follows, that an optimum is not an absolute point but changes with the duration of the observations.

The influence which the process undergoes after a length of time at a higher temperature acts as "limiting factor". These limiting factors are discussed in detail in BLACKMAN's paper.

I have investigated the extent to which this theory is applicable to the normal respiration of seedlings and in the following pages I propose to give a preliminary account of the results obtained.

I used as measure of respiration the liberation of CO_2, which was estimated according to PFEFFER's method [3]). The temperature was kept constant within 0.2° C.; in order to determine the time required by the objects to attain the constant temperature, the course of the temperature was followed thermo-electrically in a few cases; it was not possible to do this continually in the botanical laboratory.

The seedlings were placed, as free from each other as possible, on aluminium plates arranged round an axis in the respiration vessel;

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1) G. L. C. MATTHAEI, Philosophical Transactions, vol. 197 B, p. 47, 1905.

2) F. F. BLACKMAN, Annals of Botany, vol. XIX, p. 281, 1905.

3) W. PFEFFER, Untersuchungen aus dem botan. Institut zu Tübingen, Bd. 1, p. 636—645, 1885.

in this vessel there were always also two little basins of water, as humidity was found to have a great influence.

The titration of the baryta water was carried out with HCl, with phenolphthalein as indicator.

Generally I made observations at intervals of 5°; in most cases an experiment lasted 6 hours. A difficulty, which cannot be obviated, is that, when seeds are kept for a long time, the respiration is very different at different periods. The figures thus become much less comparable; in the following table I therefore give a series of experiments which were made within as short an interval as possible.

Most of the experiments were made with Pisum sativum; these will be considered in greatest detail. For every experiment 100 seedlings were used, which had germinated for one day in water and for two days in saw-dust.

Temp.	mgr. CO_2 liberated during						Average per hour
	1st hour	2nd hour	3rd hour	4th hour	5th hour	6th hour	
0°	4	4	3.6	4	3.8		3.9
5°	6.7	6	5.6	5.7	5.8		6.1
10°	12	11.2	11.1	11.4	11.4	11.3	11.4
15°	18.6	19	19.4	19.4	20	20.2	19.4
20°	28.6	30.3	30.3	30.6	30.4		30
25°	43.3	42.4	42.2	41.7	40.7	40.9	42
30°	51.7	50.9	52.2	53.6	53.5	53 5	52.6
35°	68.7	62.8	60.1	61.7	60.9	60.9	
40°	73.3	55.2	49	45.3	43	41.2	
45°	73.5	48.4	41.9	35.9	31.9	28.6	
50°	74	38.8	17.8	12	8	5.9	

It is already clear from the averages between 0° and 20° that the proportion for any rise of temperature of 10° lies between 2 and 3; from the mean of *all* the observations I find the following relation: [1])

$$\frac{A_{10}}{A_0} = \frac{10.3}{4.3} = 2.4 \quad \frac{A_{15}}{A_5} = \frac{18.5}{5.9} = 3.1 \quad \frac{A_{20}}{A_{10}} = \frac{29.1}{10.3} = 2.8 \quad \frac{A_{23}}{A_{13}} = \frac{40.3}{17.8} = 2.2.$$

The rule of VAN T' HOFF-ARRHENIUS holds good therefore up to 20°,

[1]) A_0 means respiration at 0°, etc.

with a coefficient of about 3. The observations at 0° are the least reliable, for here the quantity of CO_2 is smallest, so that the error of the apparatus, which amounts to about 0.7 mg., has the greatest influence. I have, however, not thought it advisable to make a correction for this error, as the individual differences of the plants used are much greater.

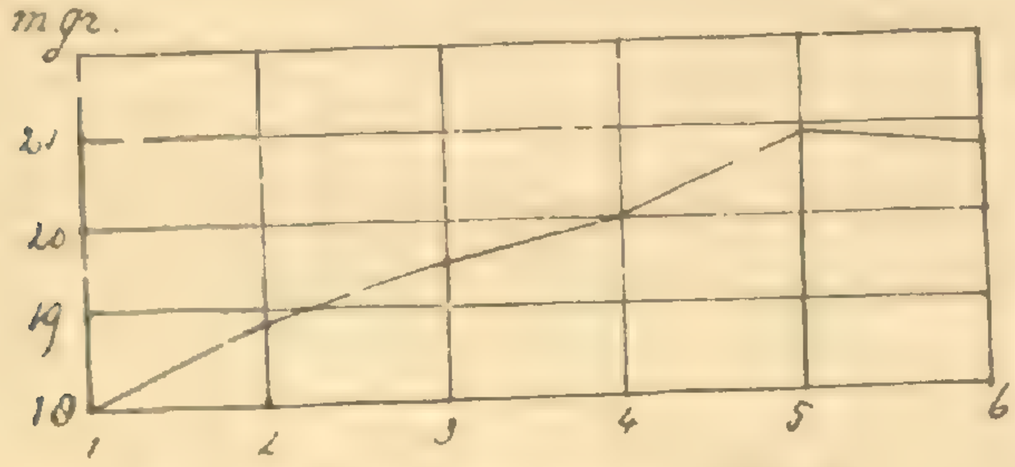

Fig. 1. Respiration at 15° C.

at 30° show an irregular course;

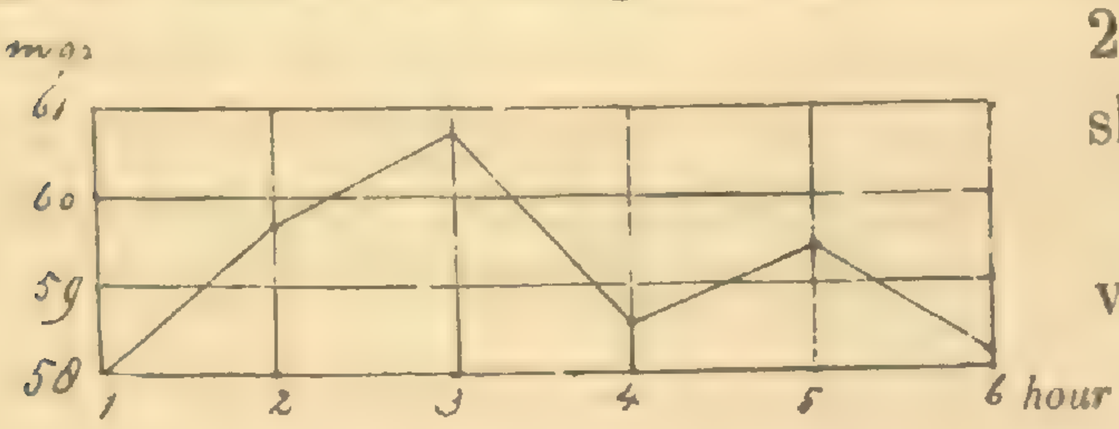

Fig. 2. Respiration at 30° C.

It is evident from the figures that at 0°, 5° and 10° the respiration is constant in successive hours; between 15° and 20° something of a rise may frequently be noted. (Fig. 1).

The curves at 25° and especially the figures quoted in the table for 25° decrease somewhat; others show the irregularity more.

Fig. 2 represents this irregular variation at 30°. We may here remark that at 30° growth is optimal and can for instance be readily detected with the naked eye after 6 hours.

At 35° respiration falls off pretty rapidly during the first hour and afterwards continues to oscillate more or less; on the other hand the falling off persists at 40°, 45° and 50°; the curves somewhat resemble logarithmic ones (fig. 3) although, as far as I have been able to ascertain, they do not satisfy such a condition mathematically. The absolute and the percentage decline becomes larger with rise of temperature. Between 35°—50° therefore a harmful factor makes its appearance, so that at 25° and 30° there is neither a rise nor a decline whereas a rise may be detected at 15°—20°.

Notwithstanding BLACKMAN's theory, there is some point in investigating the optimum of respiration, because it has been a point of dispute between various observers. The principal records are those of KREUSLER [1]) and of CLAUSEN [2]).

KREUSLER found no optimum for branches of Rubus; the respiration rises to the temperature limit of life, and this is the view which has been accepted in most text books. The highest temperature observed was 46°.6 C.

CLAUSEN found for Lupinusluteus an optimum at 40°. I wish to

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[1]) U. KREUSLER, Landwirtsch. Jahrbücher, Bd. 16, p. 711, 1887.

[2]) H. CLAUSEN, Landwirtsch. Jahrbücher, Bd. 19, p. 893, 1890.

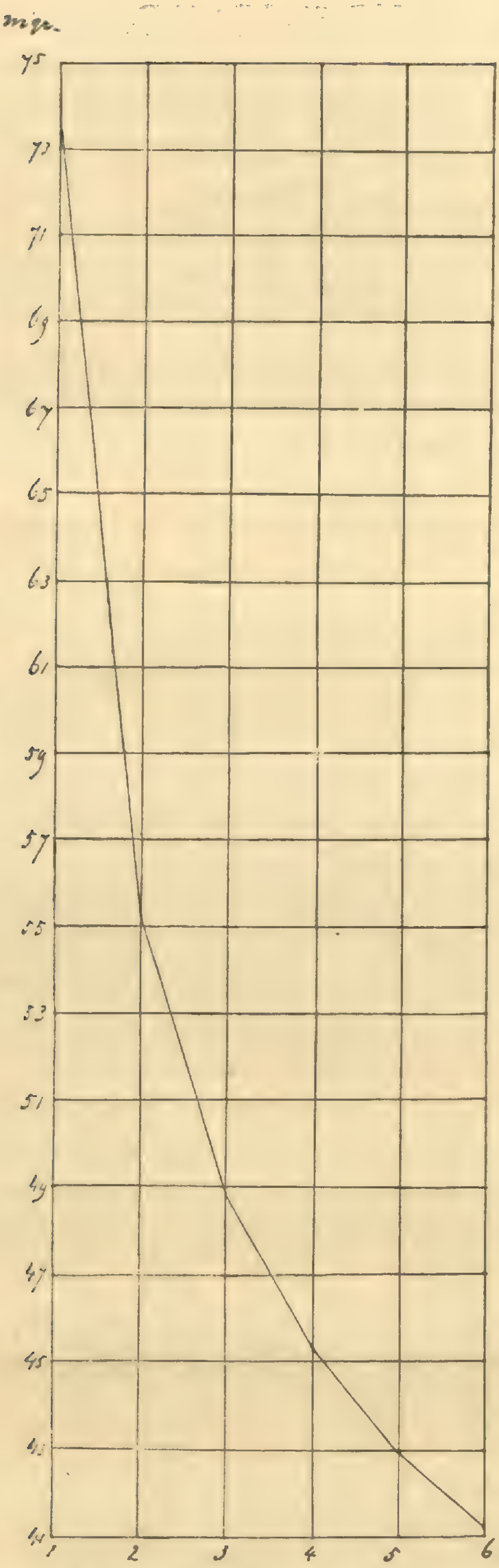


Fig. 3. Respiration at 40° C.

point out that CLAUSEN kept his seedlings for 2 hours at 50° before making an observation. The following series results from my own observations, using the figures for the 1st hour at temperatures of 35°—50°:

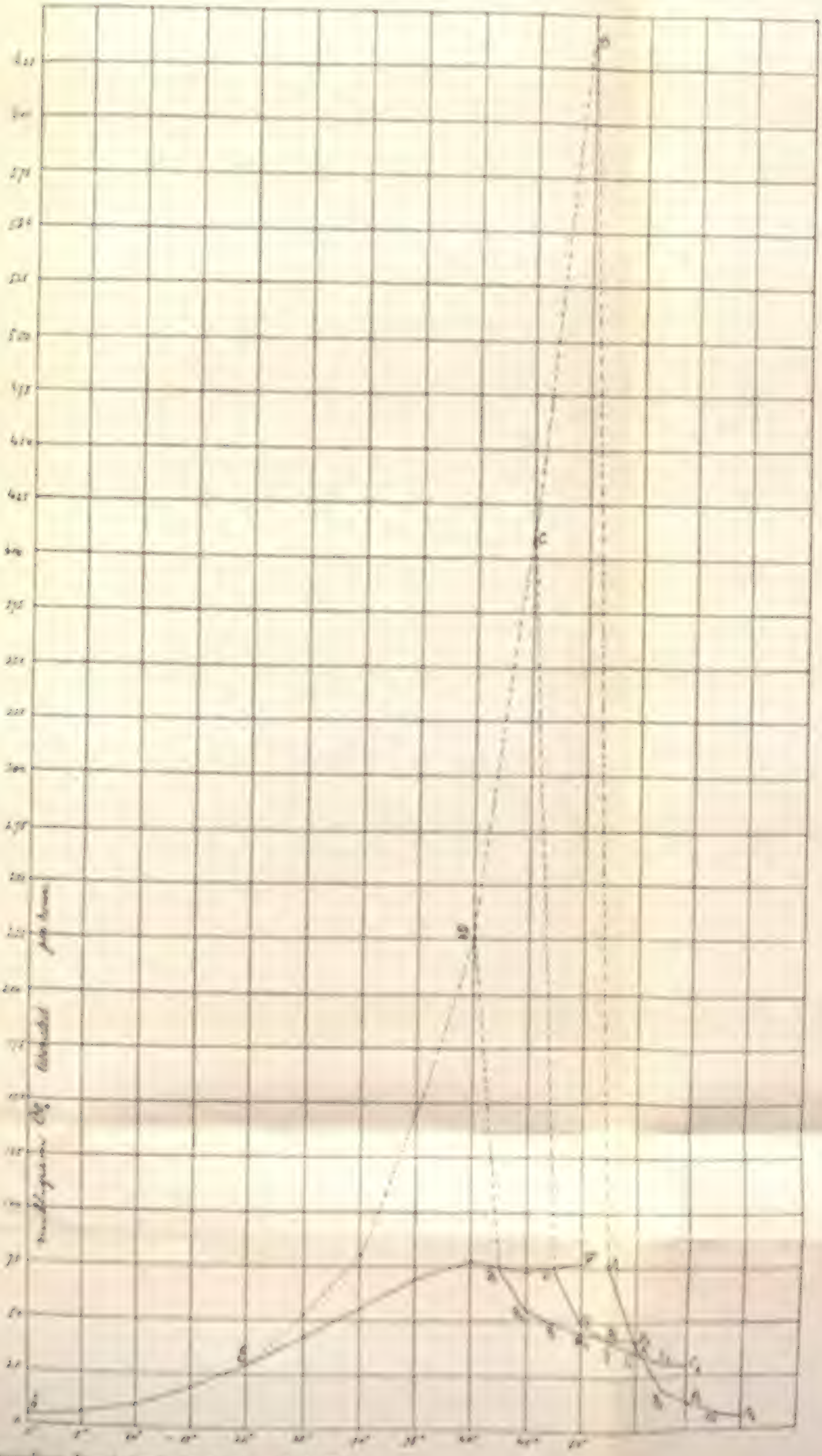
0°	5°	10°	15°	20°	25°
4.3	5.9	10.3	18.5	29.1	39.5
30°	35°	40°	45°	50°	
56.1	68.9	77.7	73	76.4.	

Thus there is obviously no question of a definite optimum; from 40° upwards the values oscillate somewhat. As is evident from the table on p. 220 the observations during the 2nd hour give a very definite optimum at 35°; on the other hand the following table, in

Temp.	CO_2 liberated per hour in mg. calculated according to observations of		
	1 hour	½ hour	¼ hour
40°	77.7	79.2	82
45°	73	82	91.6
50°	76.4	92.6	100

which the respiration at 40°, 45° and 50° has been calculated from observations of the 1st half hour and 1st quarter of an hour, shows

Fig. 5.



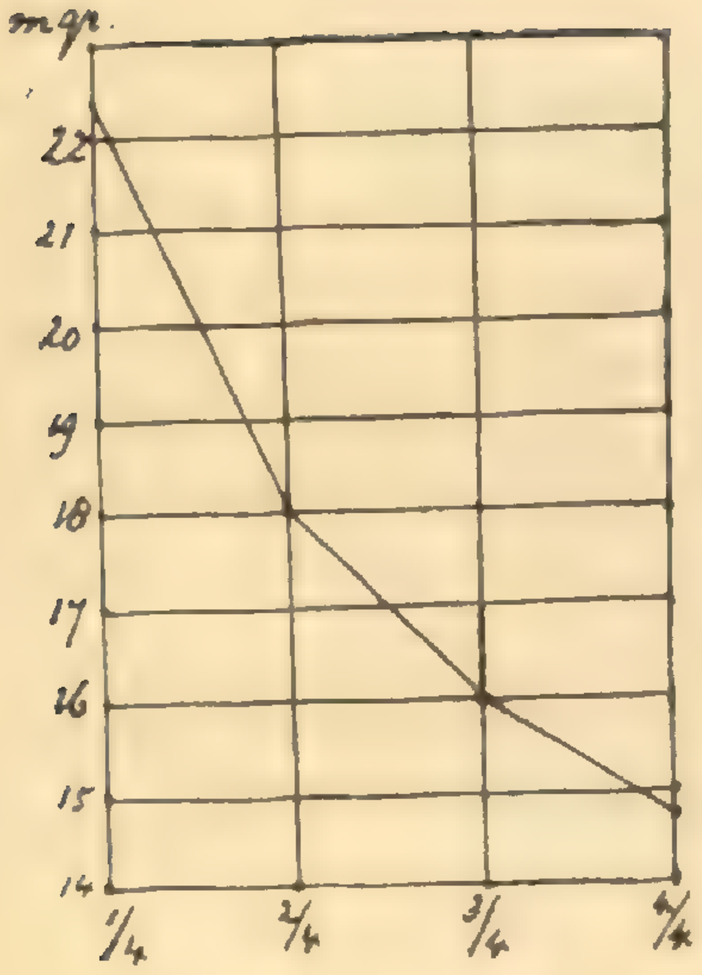


Fig. 4.
Respiration at 45° C. in successive quarters of an hour.

clear an optimum which coincides with the maximum. The observations during 4 successive quarters of an hour very clearly indicate the logarithmic course of the falling-off, as is shown by fig. 4.

We see from these figures that BLACKMAN'S view as regards the optima, is also completely applicable to the respiration of Pisum.

It remains to be considered whether BLACKMAN'S theory can be applied still further, i.e. whether the value calculated according to the rule of VAN T' HOFF-ARRHENIUS, agrees with that found by exterpolation.

Fig. 5 gives a representation agreeing completely with that of BLACKMAN [1]). The abscissa indicates the temperature, the ordinate the mgr. CO_2 liberated per hour. The line AF gives the figures found experimentally during the first hour of observation, while AB has been drawn by taking 2.8 as coefficient for a 10° temperature interval and considering the figures for 10°, 15° and 20° as standards. The relation between the amount of CO_2 and the time would have to be indicated in a second plane at right angles to the abscissa. This plane has been imagined folded over to the right, for 40° over the line drawn at 40° perpendicular to the abscissa, and further for every temperature over a subsequent line, as the points D, C, and B always have an abscissa $= 0$ in this new system. From the point 40° onwards the abscissa indicates the time after which each observation was made.

The values D_1—D_6 etc. have been taken from the table on p. 220 The lines BB_6, CC_6 and DD_6 are not smooth curves, so that the points B, C, and D may not be regarded as obtainable by exterpolation. On the other hand the greater or less inclination of the intervals BB_1, CC_1, and DD_1 agrees with the greater or less inclination of the intervals B_1B_6, C_1C_6, and D_1D_6.

The general impression which this diagram gives, is, in my opinion, that BLACKMAN'S view holds good. Nevertheless, I consider that an important deviation is found in the impossibility of deducing the theoretical points from the other figures by exterpolation. A better analysis of the whole process by which CO_2 is set free, will perhaps some day explain this deviation.

As was pointed out above, a falling-off is noticeable from 35°

[1]) BLACKMAN, l.c. p. 284.

onwards, and this must be attributable to some harmful influence. I supposed that this influence was already observable at 30° and therefore tried to show it in another way. I made a series of experiments in which I first observed the peas at 20°, then warmed them to a higher temperature and again observed them at 20°. The observation times were of course chosen in such a way that the peas could properly regain the temperature; first of all it was ascertained whether a change of temperature as such had any influence, by making experiments at 5°—25°—5°. No such influence was found; my results agree completely with those of ZIEGENBEIN [1]) in contradistinction to those of ZALENSKI [2]) and PALLADIN [3]), who ascribe a powerful influence to changes of temperature.

Now in these experiments I found a falling-off, in consequence of the harmful effect of heating to 40° and higher; this falling-off was very small after warming to 35°, so small that the effect was doubtful; an exposure to 30° did not produce any decline at all. In this way also therefore no conclusion could be reached. It was indeed found by warming for various periods to 43°, that the decline for 15 min., $^1/_2$ hour, 1 hour and $1^1/_2$ hours, when plotted, gave a logarithmic curve. The slowing of the respiration by warming to 40° was found to have disappeared completely after 8 hours.

After this I extended the experiments to Triticum vulgare and to Lupinus luteus. I only investigated the respiration in successive periods at temperatures of 0°—50°. The seedlings were treated as in the case of Pisum; they were, however, left one day longer on saw dust.

For Lupinus luteus I obtained the following results for 100 seedlings. (See following table p. 225).

It results from these figures:

That the coefficient for a temperature interval of 10° is 2.6; on determining the ratio $\frac{A_{25}}{A_{15}}$ by taking for A_{25} the respiration in the first hour, we find 2.4; by taking the mean of my other data this even becomes 2.5. The rule of VAN 'T HOFF-ARRHENIUS seems to apply here up to 25°. At 25° a certain decline is already noticeable, and at 40° this shows a logarithmic curve.

In general Lupinus is less resistant to a high temperature than Pisum; after 6 hours at 45° the turgescence of the roots had partially disappeared. As regards BLACKMAN's theory, Lupinus gives the same general result as Pisum.

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1) E. ZIEGENBEIN, Jahrbücher für wissensch. Botanik, Bd. XXV, p. 563, 1893.
2) W. ZALENSKI, Abstract in Bot. Centralblatt, Bd. 95, p. 251, 1904.
3) W. PALLADIN, Revue générale de Botanique, t. XI, p. 241, 1899.

Temp.	mgr. CO_2 liberated during						Average p. hour
	1st hour	2nd hour	3rd hour	4th hour	5th hour	6th hour	
5°	5.2	5.4	5	4.5	4.5		4.9
10°	8	7.8	7.8	8.2	8.5	8.2	8.1
15°	11.2	12.2	13.4	13.4	13.6	13.2	12.9
20°	20.9	21.9	22.9	22.5	21.1	21.5	21.8
25°	30.5	31.6	30.5	29.9	28.3	27.9	
30°	37.9	37.1	34.7	34.6	32 4	31.8	
35°	43.8	43.3	42.1	41.4	41	39.3	
40°	58	49.7	45.1	41.8	42.3	42.1	
45°	57.1	42.3	35.8	34.9	34	31.4	
50°	49.9	12	5.7				

I here append the figures obtained for Triticum vulgare, with 50 grams of 4 days-old seedlings.

Temp.	mgr. CO_2 liberated during						Average p. hour
	1st hour	2nd hour	3rd hour	4th hour	5th hour	6th hour	
0°	4.4	4.4					4.4
5°	7.6	7.2					7.4
10°	11	11.2					11.1
15°	21.3	22	22	22.9	22.4	22.4	22.1
20°	26.9	27.3	27.1	28.1	28.7		27.1[1]
25°	36.4	36.4	36.4	39.2	40.8	40.8	36.4
30°	45.7	46.1	47.3	50.3	49.7	50.5	
35°	48.2	44.6	49.7	46.2	47.7	46.8	
40°	51.8	45.4	42.2	38.2	36.4	35.8	
45°	48.7	40.4	33.7	29.7	26.7	24.8	
50°	42.2	22	12.4	10	9.2	8.8	

From *all* the available figures I find:

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[1]) The averages for 20° and 25° have been calculated from the first three hours.

$$\frac{A_{10}}{A_0} = \frac{11.4}{4.4} = 2.6 \quad \frac{A_{15}}{A_5} = \frac{20.2}{8.2} = 2.5 \quad \frac{A_{20}}{A_{10}} = \frac{27}{11.4} = 2.4.$$

Hence the coefficient is here on the average 2.5. If we consider the course at every temperature, we find that up to 30° a very distinct rise is observable; I attribute this partly to the increase of respiration with the advance of the period of germination of the plant. This increase was already observed by RISCHAVI [1]); I myself found at 20° for instance after 4 days' germination 28 mg., after 5 days in the same plants 36 mg.

At 35° the curve is very irregular, while at a higher temperature the logarithmic decline again shows itself.

In this case also the general aspect is the same as that presented by Lupinus and Pisum. The difference between the three plants is in the temperature at which the rise, the oscillations and the decline occur.

		Lupinus	Pisum	Triticum
Rise	observable at	15°—20°	20°	30°
Oscillations	„ „	20°—25°	30°	35°
Decline	„ „	25°—30°	35°	40°

It will be seen that in this respect the three plants form a series. I imagine that this different behaviour is caused by, or is at least closely connected with the reserve food which the plant has at its disposal. There is found for instance in: [2])

	Lupinus	Pisum	Triticum
as reserve food	no starch protein 37 %	starch 54 % protein 22 %	starch 74 % protein 12 %

If follows from these figures that the temperature at which the harmful effect arises, is higher when the starch content is higher, and lower in proportion as the protein content is higher. Probably there is here an indication that the course of respiration depends on the presence of proteolytic or diastatic enzymes; the falling-off at a

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[1]) L. RISCHAVI, Landwirtsch. Versuchsstationen, Bd. 19, 1876.

[2]) These data have been taken from HARZ, Landwirtschaftliche Samenkunde, 1885.

lower temperature in the seeds with a higher protein content is perhaps connected with the fact that the optimum for proteolytic generally lies lower than for diastatic ones.

In conclusion I mention a few experiments with capitula of Crepis biennis. I only made observations from 30°—50°; at each of the temperatures employed a falling-off was observable, which was very marked and gave an almost straight line. The experiments are too few in number to allow of a definite conclusion being drawn but the marked decline is in this case also most probably influenced by a want of respiratory material. According to the observations the respiration increases during the first hour right up to 45°, while the temperature limit of life is only a very little higher; after a 6 hours' exposure to 45° the flowers were dead.

My results are therefore as follows:

1. the theory of BLACKMAN is also applicable to respiration to the extent that:

a. the rule of VAN 'T HOFF-ARRHENIUS holds good from 0° to 20°—25°.

b. the optimum may be displaced with the time of observation.

c. the falling off at 40°, 45°, and 50° has a logarithmic course in seedlings.

2. the course of respiration depends very much on the nature of the reserve food.

Utrecht, Botanical Laboratory.

Chemistry. — "*On retrogressive melting-point lines.*" By Prof. A. SMITS. (Communicated by Prof. J. D. VAN DER WAALS).

(First Communication.)

Applying the method indicated by VAN DER WAALS [1]) we find for the melting-point line under the vapour pressure the following equation:

$$\frac{dx_L}{dt} = -\frac{1}{T}\left(\frac{\partial x_L}{\partial p}\right)_T \frac{(x_L - x_g)\{(V_g - V_L)(Q_m + Q_{SL}) - (V_L - V_S)Q_{LG}}{(V_g - V_L)\{x_L - x_g)V_S - (x_S - x_g)V_L + (x_S - x_L)V_g\}}$$

in which Q_{SL} = mol. heat of melting of a compound of the concentration x_S;

Q_m = differential mol. heat of mixing of the melted compound with a solution of the concentration x_L;

Q_{LG} = mol. heat of evaporation of a solution of the concentration x_L.

[1]) Versl. Kon. Akad. v. Wet. 28 Febr. 1885 and Rec. Trav. chim. **5**, 336 (1886).

x_S, x_L and x_g denote the concentration of the coexisting solid, liquid, and vapour phase, expressed in gr. mol. of the component B per one gr. mol. total, and V_S, V_L, and V_g indicate the mol. volumes of these phases.

If we now confine ourselves to the system $H_2O - Na_2SO_4$, and if we apply the above formula successively to the melting-point lines of Na_2SO_4 10 aq, of rhombic Na_2SO_4, and of monoclinic Na_2SO_4, we arrive at remarkable results.

Starting with the melting-point line of Na_2SO_4 10 aq, we must remark beforehand that for the case that no maximum or minimum occurs in the p-x-figure, which is here excluded, $\left(\frac{\partial x}{\partial p}\right)_T$ is always negative.

As now the denominator is already positive some degrees below the melting point in consequence of the predominating value of $(x_S - x_L)V_g$, the sign of $\frac{dx_L}{dt}$ will be further governed by the numerator, and so we see that it follows from the positive values of $\frac{dx_L}{dt}$ for the melting-point line of Na_2SO_4 . 10 aq that the numerator, so the sum of the caloric quantities, is positive there.

This is the usual case, and does not call for further remarks.

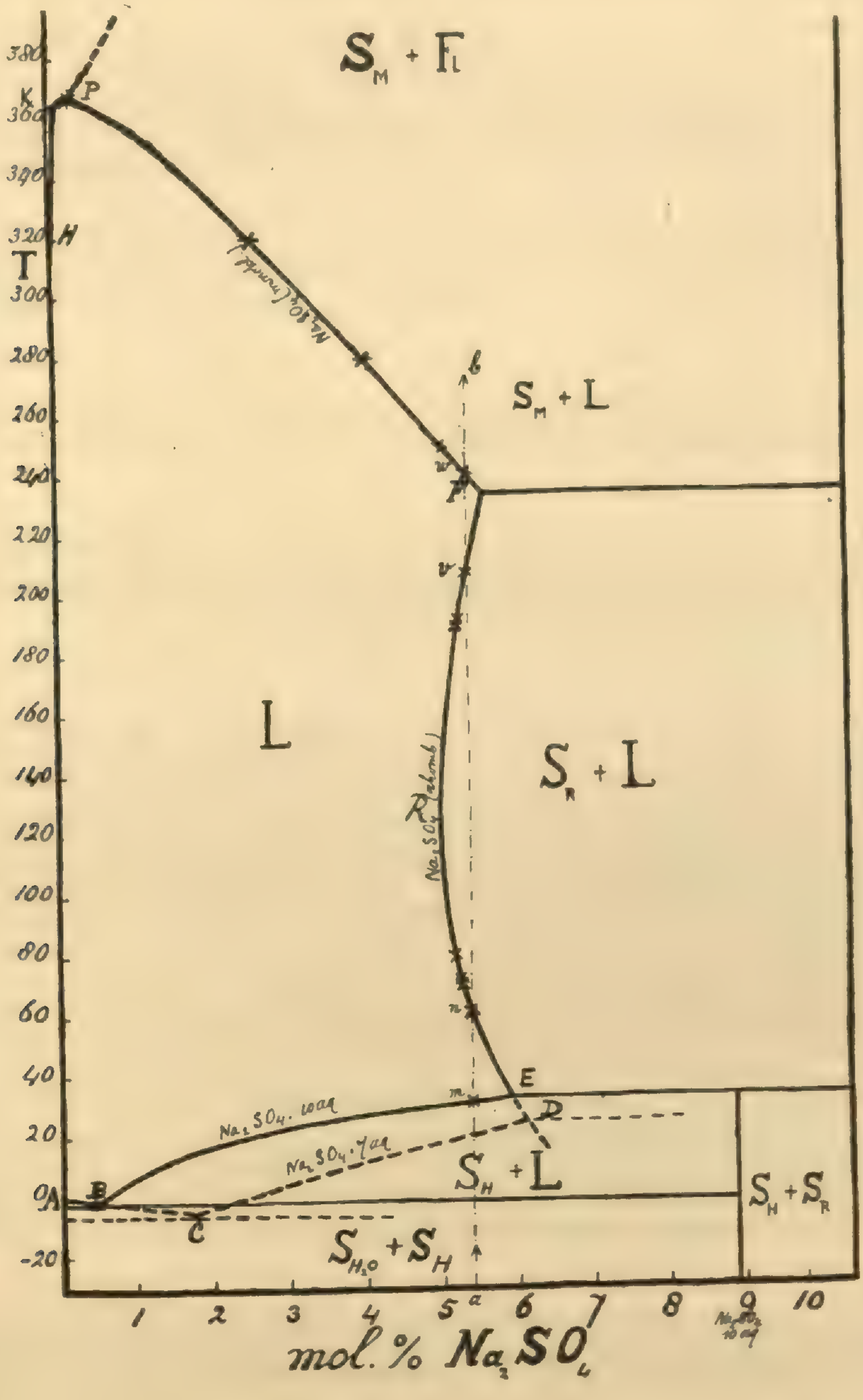
From the figure p. 229, which represents the projection of the three-phase regions $S_B + L + G$ on the T,X-plane we see, however, that the melting-point line of the rhombic Na_2SO_4, which becomes stable in E presents the particularity of at first retrograding, and now the question suggests itself, how this behaviour can be accounted for.

When in a solution the phenomenon of hydration takes place, the differential heat of mixing, which is on the whole thermodynamically positive, can become negative, because the formation of hydrate is usually attended with generation of heat.

Now it is clear from the above equation that the melting-point line can become a retrogressive one only when the numerator with the caloric quantities has become *negative*, which will be the case when Q_m assumes a predominantly large *negative* value.

If we now consider the transition point E, we know that the solution, which coexists there with solid Na_2SO_4 . 10 aq., is also in equilibrium with rhombic, anhydrous Na_2SO_4. Now if this solution is hydrated, as is made probable by various phenomena, it is clear that the differential heat of mixing of anhydrous Na_2SO_4 must be much greater than that of Na_2SO_4 . 10 aq., as the molecules, before

being solved have already been hydrated to a certain extent in the latter case. In consequence of this Q_m can have become so strongly



negative that it predominates over the other positive caloric quantities, of which Q_{SL} now relates to the anhydride.

So, seen in this light, we may expect that if a hydrate splits off much water at its transition point, a retrograding melting point line will arise, because then the probability that the change in the heat of melting is smaller than that in the heat of mixing, is very great.

From the T,X-figure it appears further, that the retrogression of the melting-point line of rhombic anhydride becomes less and less

at higher temperature, so that $\frac{dx_L}{dt}$ passes through zero at the point R, after which it assumes again positive values.

This peculiar phenomenon can hardly be explained in another way than by assuming a retrogression of the hydration in solution with rising temperature, which causes the heat of mixing to become smaller negative, so that the denominator of the fraction first becomes *zero* and then again positive.

This continues like this up to the transition point F, where another remarkable phenomenon is met with. In this point, where the rhombic modification passes into the monoclinic one, which is a transformation without splitting off of water, arises a strongly retrograding melting-point line too. Now, as the difference of state between rhombic and monoclinic Na_2SO_4 disappears in the liquid state, we know with certainty that the heat of mixing of the two melting-point lines, is exactly the same in the point F, from which follows that the reversal of sign of $\frac{dx_L}{dt}$ must be attributed to the circumstance that Q_{SL} now refers to the monoclinic modification, the heat of melting of which must be considerably smaller than that of the rhombic form.

If we now proceed to higher temperature, complications appear, as we now approach the critical state, in which $V_G - V_L$ and Q_{LG} become continually smaller, till they finally become *zero* in the critical point.

In this point $\left(\frac{\partial x_L}{\partial p}\right)_T = -\infty$, so that $\frac{dx_L}{dt}$ becomes $= -\infty$.

In conclusion we may still remark that it follows from this consideration that the hydration continues to exist even at the critcal temperature.

If we now further consider that it follows from my investigation of the system ether-anthraquinone that a partly retrogressive melting-point line must exist also there, the supposition suggests itself that the whole phenomenon is in connection with the formation of compounds in solution, but it is exactly the case ether-anthraquinone that admonishes us to caution, because nothing is known of a compound between these two substances in solid state. So it seems better to me to assume that the molecules of the two substances approach each other very closely in solution, leaving undecided whether this state can be called a compound or not.

Amsterdam, Sept. 1909.

Anarg. Chem. Laboratory of the University.

Chemistry. — "*The P,T,X-spacial representation of the system ether-anthraquinone.*" By Prof. A. SMITS. (Communicated by Prof. J. D. VAN DER WAALS).

As the type ether-anthraquinone occurs so frequently, and the spacial figure of the substances belonging to this system deviates so greatly from the P,T,X-representation given by BAKHUIS ROOZEBOOM on account of the very different state of the two components at the same temperature, it seemed desirable to me to draw up a spacial representation of the system ether-anthraquinone.

The figures 1, 2, and 3 now give us a view of this P,T,X-spacial representation taken from different sides, and may contribute to facilitate the insight into the peculiarities of this system.

In fig. 1 the plane a, l, f, k, s, h, g represents the p,x-section corresponding with the critical temperature of the component ether, which is thought on the left.

So the point a indicates the critical pressure of pure ether, and the line $a\,l$ the unsaturated liquids containing anthraquinone, which coexist with the vapours on the line $a\,g$.

The three-phase-pressure line g, l, s, on which the equilibrium between vapour g, saturated liquid l, and solid anthraquinone s lies, forms the upper boundary of the two-phase equilibrium between solid anthraquinone and vapour, the vapour phases of which lie on the vapour line h, g, and the solid phases coexisting with them on hs.

The line $g\,l\,s$ is at the same time the lower boundary of the equilibrium between liquid and solid anthraquinone just as of that between liquid and vapour, and $l\,f$ or the meltingpoint isotherm indicates the liquid phases, and $s\,k$ the solid phases.

AS VAN DER WAALS showed, the lines $h\,g$ and $l\,f$ are two branches of one and the same curve, which possesses a partly metastable, partly unstable intermediate portion between g and l. If we wish to have a name for the whole curve, *solubility-isotherm* is convenient.

If we now proceed to higher temperatures, i. e. in a direction normal to the just discussed p,x-section to the left, the principal change is this that the lines la and ga pass continuously into each other, and the liquid-vapour region moves a little to the right as the lines not only get detached from the axis but the vapour and liquid points g and l move also to greater anthraquinone concentrations.

The point where the continuous transition between liquid and vapour takes place lies on the plaitpoint curve $a\,p\,q\,b$, which connects the critical point of ether with that of anthraquinone, and running on the liquid-vapour surface connects the points of maximum pressure in the different p,x-sections.

The three-phase region for $G + L + S_B$, which is formed by the successive three-phase-pressure lines $g\,l\,s$, runs at first with rise of temperature towards higher pressure, but finally descends again and terminates in the triple point of B.

As fig. 2. shows particularly clearly, the peculiarity of the system ether-anthraquinone consists in this, that the plaitpoint-curve meets the three-phase region in the points p and q.

Fig. 2, namely, gives the photograph of the spacial figure seen in the direction of the x-axis from A towards B.

The line hr is the vapour-pressure line of solid B; r is the triple point and rb the vapour-pressure line of liquid B terminating in the critical point b.

The lines gp and qr are the projections of the broken three-phase region on the P,T-plane, and between a and p, and q and b are found the two branches of the plaitpoint curve, which is metastable between p and q.

If for the sake of simplicity we confine ourselves for the present only to the point p, it is noteworthy that in this point three remarkable points coincide, viz. the plaitpoint with the vapour point g, and the liquid point l of the three-phase equilibrium $G + L + S_B$.

So in this point p a saturated solution shows the critical phenomenon, and when the mixture contains more B than corresponds with the point p, the said critical phenomenon will occur in the presence of solid B at the temperature of this point.

Exactly the same thing now applies to the second critical end point q.

Above the temperature of the point p solid B can only coexist with a fluid phase, and the peculiarly curved surface $npedqm$ indicates the fluid phases, which can be in equilibrium with solid B at different temperatures and pressures.

The upper part $pedq$ passes continuously into the melting-point surfaces $lfep$ and $qder$ (fig. 2), whereas the lower part joins continuously the vapour surfaces $hgpn$ and mqr. It is now still more apparent than before that p and q entirely agree with each other with only this difference that what is observed for p with rise of temperature, takes place for q with fall of temperature. Thus fig. 1 shows that the continuous liquid-vapour surface terminates at p, and begins again at q. It is further worthy of note that the solubility curve of B under the vapour pressure in normal cases runs towards higher concentrations of B at higher temperature.

As, however, a saturated solution becomes identical with the coexisting vapour at p, the solubility line lp, fig. 1, must continuously

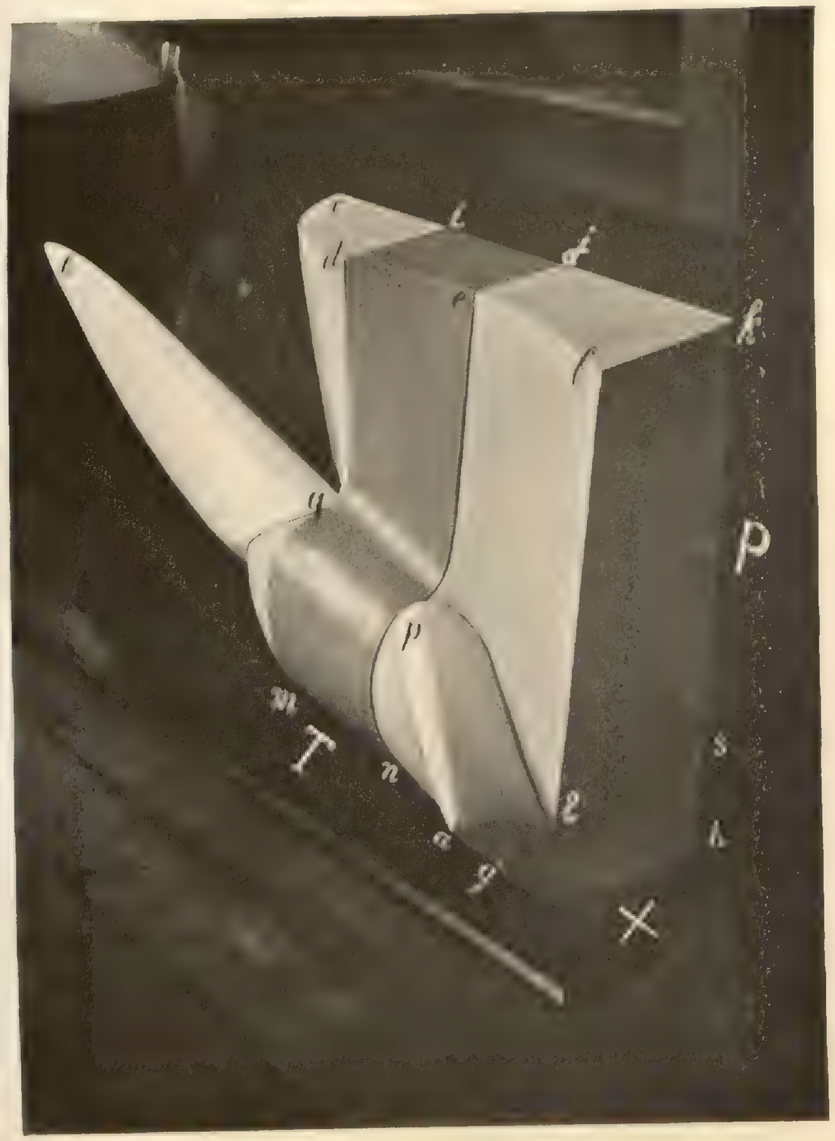


Fig. 1.

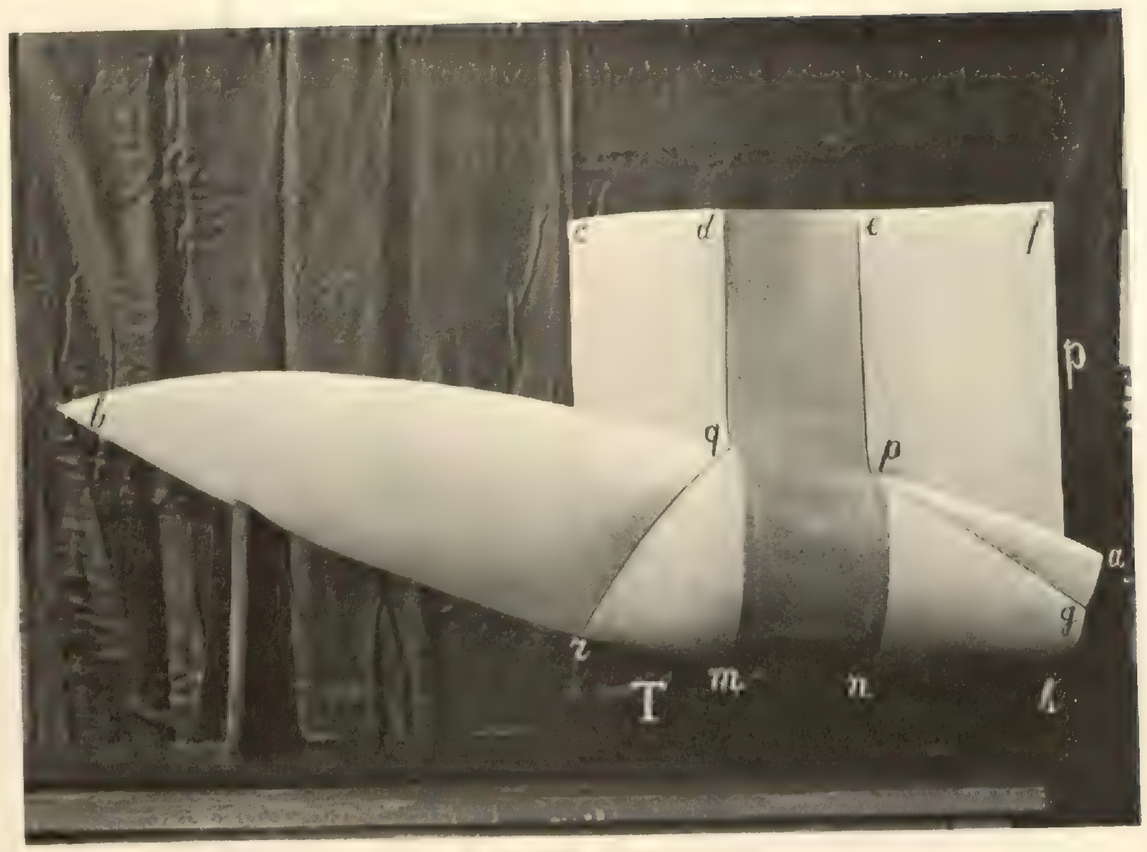


Fig. 2.

P

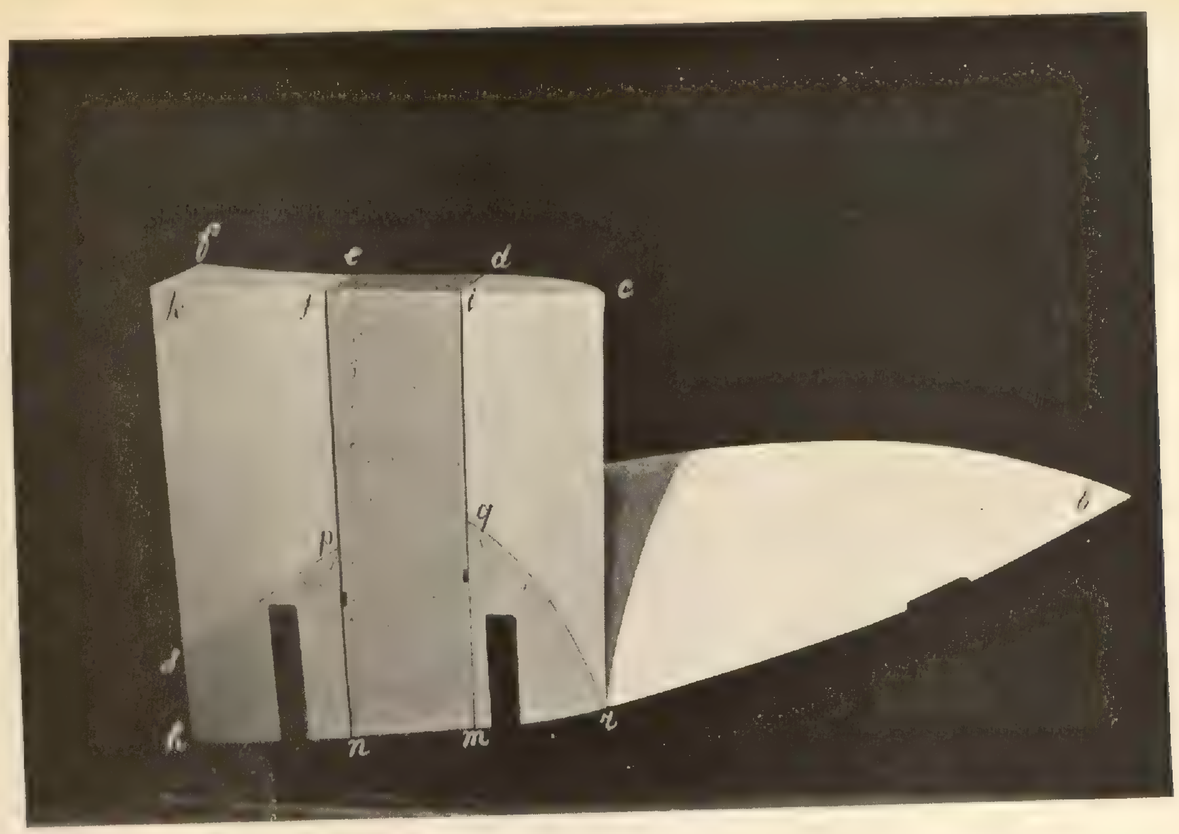


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Fig. 3.

pass into the vapour line qp in p, and in consequence of this the solubility line will have to bend to the left, which gives rise to the phenomenon of retrograde solubility.

As is evident from fig. 1 we get the retrograde phenomenon at q on the vapour line for the same reason.

Another remarkable phenomenon which fig. 1 also reveals, is that of retrograde condensation and of retrograde solidification.

It is clear that if starting from the point a we roll a vertical tangent plane along the spacial figure in the direction of the axis of temperature, this plane touches the vapour sheet of the continuous liquid-vapour surface first for some time, but a change has come before the temperature of the point p is reached. The before mentioned vapour sheet is no longer touched, and contact now takes place with the vapour sheet of solid B. The change takes place at the moment that the two vapour surfaces are touched, which reveals a *double retrograde phenomenon.*

This behaviour now proves that we have first only the phenomenon of retrograde condensation, which at a certain temperature is succeeded by the phenomenon of retrograde condensation and solidification, and afterwards only by the phenomenon of retrograde solidification.

What has been discussed here holds also for the neighbourhood of q, but here the phenomenon takes place in reversed order. Starting from q we get first retrograde solidification and then retrograde condensation with rise of temperature.

The phenomenon of retrograde solidification continues to exist between p and q (this was found for ether-anthraquinone, but it has proved not necessary theoretically), and this is the reason why the surface for the equilibrium between solid B and fluid has such a peculiar shape.

Now it is to be seen in fig. 1 that at the temperature of p the solubility isotherm has a somewhat different shape from that of q; this is in perfect harmony with the experiment. It has viz. appeared that the part of line pe running to the right is much larger than that of line qd, and in consequence of this the melting-point line will have a shape as indicated by the line $cdef$, which is a melting-point line with two vertical tangents at a pressure higher than that corresponding with the highest three-phase pressure [1]).

In conclusion I may call attention to fig. 3, which shows the backside of fig. 2, or rather the spacial figure on the B-side seen slightly from above, so that the melting-point line $cdef$ is clearly visible.

Amsterdam, Sept. 1909.

Anorg. Chem. Laboratory
of the University.

[1]) This question will be examined in detail.

Physics. — *"On the influence of the pressure on the miscibility of two liquids."* By Dr. J. TIMMERMANS and Prof. PH. KOHNSTAMM. (Communicated by Prof. J. D. VAN DER WAALS).

1. *Survey of earlier experimental investigations.*

A survey of the experiments on the influence of the pressure on the miscibility of two liquids, has often been given [1]); so we shall confine ourselves here to some short remarks.

If we leave ALEXEJEFF's experiments which were unsuccessful, out of consideration, KLOBBIE and DE KOWALSKI are to be considered as the first that have worked in this department; they succeeded in showing qualitatively the influence of the pressure on the miscibility.

Further KUENEN has carried out a systematic series of investigations, which has remained up to now the most important contribution to our knowledge on this subject. And lastly VAN DER LEE and BÜCHNER have subjected two systems of particular importance to a closer quantitative investigation, while BÜCHNER found at the same time a great number of new systems.

It appears from all these investigations that the influence of the pressure on the miscibility is slight on the whole, as it is, indeed, almost always the case in condensed systems, in which no vapour phase is present.

2. *Theoretical considerations.*

The theory of these phenomena comprises also the study of the different possible forms of the saturation curve of a binary mixture, on which two liquid phases are in equilibrium. We confine ourselves to calling attention first to ROTHMUND's supposition [2]), according to which all these saturation curves might finally be reduced to the type of a closed curve, as it is realized in the system nicotine and water; then to that of TIMMERMANS [3]), on the possibility of "negative"

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[1]) N. J. VAN DER LEE, Thesis for the doctorate, Amsterdam, 1898, and Zeitschrift für physikalische Chemie. 33, 622, 1898.

BÜCHNER. Thesis for the doctorate, Amsterdam, 1905, and Zeitschrift für physikalische Chemie. 56, 257, 1906.

J. P. KUENEN, Philosophical Magazine (6), 6, 637, 1903, and Verdampfung und Verflüssigung von Gasgemischen (BREDIGS Handbuch der angewandten physikalischen Chemie).

[2]) V. ROTHMUND, Thesis for the doctorate, München, 1898, and Zeitschrift für physikalische Chemie, 26, 433, 1898.

[3]) J. TIMMERMANS, Zeitschrift für physikalische Chemie, 58, 144, 1907, and Bulletin de la Société chimique de Belgique, 23, 129, 1909.

saturation curves, i. e. curves which would remain entirely inside the metastable region, so at pressures lower than the vapour pressure of the mixture (negative pressures); finally to the opinion of many other authors, according to whom the existence of a lower critical point would often, if not always, be the proof of a chemical action between the components (hydration), or of the presence of a third component [1]).

KUENEN (loc. cit.) has carried this investigation an important step forward by drawing attention to the fact that for the consideration of the different types of saturation curves, the vapour phase must necessarily be taken into account, as well as the two liquid phases. This enabled BÜCHNER (loc. cit.) to give a very interesting classification of the different types, which, however, is still incomplete.

Starting from VAN DER WAALS'S theory, VAN LAAR [2]) has finally tried to give an analytical solution of the problem in a long series of papers. His results are of importance particularly because they showed that under certain circumstances non-miscibility can occur for perfectly normal substances, a fact which was generally doubted at the time [3]). On the other hand a serious objection to these investigations is that starting from a special case, they represent it as the general one, and therefore must pronounce as abnormal a type occurring for normal substances, while they overlook a great many others, and further that they, very onesidedly, lay the stress on the existence of open plaits (not closed on the side $v = b$), a circumstance, which by no means can be considered as a *result*, as it immediately follows from the arbitrary, if not erroneous *supposition* of the linear dependence of b and x, as VAN DER WAALS had already shown in the Théorie Moléculaire. For the case at last that one of the components is abnormal, VAN LAAR arrives at saturation lines of very complicated form, which the experiment has never found as yet. We hope to return to this subject later on in connection with further experiments [4]).

3. *Investigation by* VAN DER WAALS. [5])

At last VAN DER WAALS resumed the investigation of the

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[1]) WL. DOLGOLENKO, Zeitschrift für physikalische Chemie. 62, 498, 1908.

[2]) J. J. VAN LAAR, These Proc. 1905 and 1906.

[3]) R. A. LEHFELDT, Philosophical Magazine, 47, 284, 1899.
VAN DER WAALS, Continuität II § 12.

[4]) In this survey we pass KEESOM and KAMERLINGH ONNES' investigations by, as these refer to a range of temperature altogether different from that of our experiments.

[5]) J. D. VAN DER WAALS, Archives néerlandaises, (2), 13, 77 and 799, 1908 and These Proc. Vol. X and XI (1907—1909) passim.

"longitudinal plait" two years ago, and owing to a new, particular, chiefly geometrical method he has arrived at a series of important results, which may be tested by experimental data; but the problem has proved to be so intricate that a treatment conclusive in all respects has not yet been reached. We mention some of the most important results.

a. The miscibility or non-miscibility of two liquids depends in the first place on the presence and situation of the locus $\frac{\partial^2 \psi}{\partial x^2} = 0$.

Non-miscibility occurs a good deal more frequently than was believed up to now, particularly at lower temperatures or in the critical region, and also when the components are perfectly normal.

b. A sharp distinction must be made between the case that a branch plait, which has crossed the binodal curve, retreats again inside it, and the case of the splitting-up of a plait, in consequence of which the branch plait leaves the main plait altogether. Both cases may be found in an upper mixing point (BÜCHNER's upper critical end point), but in one case an increase of pressure promotes the miscibility, in the other it diminishes it. KUENEN, in his above-mentioned paper, had already pointed out this difference, without, however, more closely investigating the circumstances on which this difference depends, and which cause either one or the other phenomenon to appear.

c. The case of the splitting up of a plait is only possible when in the system also a minimum critical temperature and a maximum vapour pressure occur.

A plait can retreat both when in the vapour-pressure line a maximum occurs, and when this is not the case.

d. When the region of non-miscibility extends as far as in the critical region proper (vapour-liquid) of the system, continuity takes place of the plaitpoint-line proper and the plaitpoint-line of the "longitudinal plait", in consequence of which rather complicated forms may originate.

e. KORTEWEG's [1]) general theory of plaits has shown that a plaitpoint can only disappear from the surface either by coincidence with another plaitpoint or by leaving the surface along one of the boundary lines $x = 0$, $x = 1$ (critical point of the components), or by leaving the surface through the line $v = b$. In this case we have to do with an "open" plait [2]), in that sense that even infinite

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[1]) Archives néerlandaises, I, 24, 295. 1889.

[2]) In physical sense; it is not an open plait in KORTEWEG's sense, Arch. Néerl. XXIV p. 5.

pressure does not bring about mixing. Whether such "open" plaits occur is in no way to be decided at the present state of our knowledge. *Theoretically* not because the supposition that b depends linearly on x leads to the existence of an open plait, but the supposition that b is a quadratic function of x [1]), on the other hand, excludes the possibility of such plaits. Now it is certainly true that both these suppositions are wrong, because they do not take account of the variability of b, and as long as we do not know the form of this variability it is certainly not possible to decide which of the simple suppositions mentioned agrees most closely with the truth. *Experimentally* not because then not only excessively high pressures would be required, but moreover in most cases the appearance of the solid state renders any investigation impossible. Hence whether open plaits are possible or not will be left entirely an open question in what follows; in our figures, where necessarily either the one or the other supposition had to be made we shall assume that open plaits do not occur, because in this way we get the simplest and clearest figures, which are then easily modified in such a way that they hold for the opposite supposition.

4. *Purpose of the investigation.*

In this first investigation we propose to examine a great part of the binary mixtures, of which it is known that they possess a critical point of mixing within easily attainable limits of temperature, for so far as is necessary to determine to what category they belong in a simple classification, which gives a survey of the possible cases. Our attention was chiefly directed to those systems that consist of two normal substances and for which also the occurrence of chemical compounds may be considered as excluded; but we have also examined other systems of very different composition in the hope of finding instances of all those types that are predicted by the theory. This investigation, which must necessarily be rather cursory on account of the great many systems which are to be examined, will be followed by accurate measurements on some of the systems which seem to us the most interesting.

The substances used were partly taken from the store of the Laboratory, partly bought at KAHLBAUM'S; none of them have been purposely purified, but the comparison of the critical end-points which we found with those given by others, proves that the purity

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[1]) With the well-known value of b_{12}.

of our substances was in general sufficient for the purpose in view. Yet it is useful just to consider what might be the influence of impurities on the extent and the sense of the change of the plaitpoint temperature with the pressure. We expect a priori that this influence will be small, unless we should just happen to be in the neighbourhood of a maximum or minimum of plaitpoint temperature; in this case the influence of the pressure itself is very small, and even traces of impurities might modify the sense of the observed changes.

The following examples, chosen as unfavourable as possible, show: the first the slight influence which the most common contamination (water) has on the value of dt/dp, the second how in a special case a trace of impurity has already sufficed to change the sign of this differential quotient.

	Kr. T.	$\frac{dt}{dp}$
I. Methyl alcohol and cyclohexane	59.0	+ 0.03
do contaminated with water	92.8	+ 0.06
II. Methylalcohol and methylene iodide, contaminated with traces of iodine	93.8	— 0.004
after a reaction between the free iodine and the mercury of the Cailletet-tube, in which green mercuroiodide is formed	82.9	+ 0.008

5. *Method.*

As in our present investigation we only purposed to determine the direction and the order of magnitude of the change of the plaitpoint temperature with the pressure, the method used was simplified as much as possible with a view to the quickness of the measurements. Mixed in the proper proportion the substances to be investigated were put in a Cailletet tube, which was provided with a long capillary; after the air had been expelled as thoroughly as possible, the capillary was sealed. The tube was surrounded by a jacket filled with water or paraffin, which was brought at the required temperature by an electrical heating apparatus; the bath was stirred by the hand; the temperature was read on a GEISSLER thermometer, graduated in degrees, and the pressure was determined by a manometer of SCHÄFFER & BUDENBERG divided into atmospheres. Of course the tube was provided with an electro-magnetic stirrer, and in order to reach the state of equilibrium as easily as possible, the readings

I	II	III	IV	V	VI	VII	VIII
1. Nitrobenzene + n-Butane	—154°	—	28°,3	15—100	−0°,04	Not the crit. concentration.	I
2. " + Isopentane	—188	22° Konovalow	32 ,6	1—140	−0 ,04		I
3. " + Hexane	—235	15 Bingham	17 ,25	1—150	−0 ,015	No vapour pressure Maximum.	I
4. " + Decane diiso	—330	27 ,5 Bingham	29 ,0	5—120	−0 ,005		I
5. o-Nitrotoluene + Isopentane	—188	—	9 ,5	1—150	−0 ,025		I
6. Aniline + n-Butane	426°—154	—	83 ,0	15—90	−0 ,08		I
7. " + Hexane	426 —235	65 Bingham	63 ,8	5—155	−0 ,012	No Maximum.	I
8. " + Decane diiso	426 —330	82 Bingham	82 ,0	5—180	−0 ,001		I
9. " + Amylene	426 —201	19 Konovalow	21 ,8	1—165	−0 ,02		I
10. " + Cyclohexane	426 —280	—	31 ,3	1—60	+0 ,005		IIa
11. m-Toluidine + Decane diiso	429 —330	32 Bingham	38 ,5	10—75	−0 ,003	No Maximum.	I
12. Methyleniodide + Hexane	—235	—	105 ,4	5—155	−0 ,02		I
13. " + Decane diiso	—330	90 Bingham	119 ,5	5—65	−0 ,015	The methyleniodide was coloured by free iodine.	I
14. " + Cyclohexane	—280	—	34 ,5	5—120	+0 ,01		I
15. Sulphur dioxide + Hexane	156 —235	14 Bingham	11 ,9	10—130	+0 ,023	The observations were rendered difficult by the slight difference of the indices of refraction.	IIa
16. " " + Decane diiso	156 —330	32 Bingham	34 ,1	10—130	+0 ,03		IIa
17. Chrysene + Ether L	—195	207 Büchner	218	49—61	+0 ,75	No Maximum. No Maximum.	III
18. Carbonic acid + Diphenylamine L	31 —	40 ,1 Büchner	—	122—162	+0 ,15	It is impossible to reach the lower critical point.	III
19. Ethane + Methylalcohol L	34 —240	see Kuenen l. c.	—	—	—	No Maximum.	III of V?
20. " + Ethylalcohol L	34 —243	31 ,9 Kuenen	—	—	+1 ,02		III
21. " + n-Propylalcohol L	34 —264	38 ,7 Kuenen	—	—	+0 ,94	No Maximum.	III
22. " + Butylalcohol L	34 —265	38 ,1 Kuenen	—	—	+1 ,10		III
23. Methylalcohol + Propane	240 —102	21 ,15	—	10—100	−0°,12 to −0°,04	A vapour pressure Maximum.	IIb
24. " + Isobutane	240 —145	—	20 ,1	10—140	0 to +0°,008	The observations were rendered difficult by the slight difference of the indices of refraction.	IIba
25. " + n-Butane	240 —154	—	16 ,6	20—150	+0°,007		IIa
26. " + Isopentane	240 —188	19 ,4 Kuenen	—	1—100	+0 ,02	A Maximum.	IIa
27. " + Hexane	240 —235	37 ,0 Kuenen	—	1—264	+0 ,03		IIa
" + "	240 —235	42 ,4 Rothmund	42 ,2	1—105	+0 ,032		IIa
28. " + Decane diiso	240 —330	76 Bingham	86 ,8	10—150	+0 ,04		IIa
29. " + Cyclohexane	240 —280	47 Lecat (1)	59 ,0	5—120	+0 ,03	A Maximum.	IIa
30. " + Carbon disulphide	240 —273	40 ,5 Rothmund	48 ,5	5—85	+0 ,015		IIa
31. Methyleniodide + Ethylalcohol	—243	22 Bingham (?)	93 ,8	5—75	−0 ,004	Coloured distinctly red by free iodine.	I
" + " (after reaction)	—243	—	82 ,9	5—205	+0 ,008	Green iodide was formed by reaction between free iodine and mercury; the red colour disappeared.	IIa
32. " + Isopropylalcohol	—235	—	93 ,2	1—200	+0 ,012		IIa
" (after reaction)	—264	—	75 ,7	1—100	+0 ,006		IIa
33. " + n-Propylalcohol (no reaction)	—265	—	77 ,5	1—80	+0 ,012	No reaction.	IIa
34. " + Isobutylalcohol (no reaction)	—240	—	62 ,0	5—195	−0 ,025		I
35. Brom-naphtaline (α + β) + Methylalcohol	—265	—	8 ,6	1—180	−0 ,01		I
36. " + Isobutyl "	—322	—	42 ,4	5—210	+0 ,025		IIa
37. " + Acetic acid	360 —	—	106 ,6	5—65	+0 ,035		IIa
38. Chlorbenzene + Formic acid (anhydrous)	289 —	—	82 ,0	5—65	+0 ,03	A Maximum.	IIa
39. Benzene + Formic acid	330 —235	—	18 ,0	1—90	+0 ,01		IIa
40. Decane + Acetone	330 —286	40 Bingham	55 ,1	1—150	+0 ,016	Slight difference of the indices of refraction.	IIa
41. " + Propionitrile	—243	31 Schreinemakers	24 ,3	10—160	−0 ,005		I
42. Ethylenecyanide + Ethylalcohol	—265	—	67 ,0	5—155	−0 ,004		I
43. " + Isobutylalcohol	—239	110 Rothmund	109 ,10	10—170	−0 ,03		I
44. Resorcin + Benzene	—346	—	151 ,5	5—65	−0 ,025		I
45. " + Ethylbenzene	31	25 Büchner	24 ,8	83—127,5	+0 ,20	No Maximum.	III
46. Carbonic acid + o-Nitrophenol L	364	113 ,6 Alexejeff	116	5—125	−0 ,07	A Maximum.	I
47. Water + Second. Butylalcohol	364 —265	130 Alexejeff	134 ,2	10—180	−0 ,03	Slight difference of the indices of refraction.	I
48. » + Isobutylalcohol	364	151 Rothmund	137 ,7	1—155	−0 ,065	Not the crit. concentration.	I
49. " + Methylethylketone	364	24 ,3 Rothmund	23 ,2	1—195	−0 ,06	A Maximum.	I
50. " + Isobutyric acid	364	—	95 ,0	5—60	−0 ,03		I
51. " + Isovaleric acid	364 —286	113 ,2 Rothmund	111 ,0	5—165	−0 ,02	Slight difference in the indices of refraction.	I
52. " + Propionitrile	364	55 Schreinemakers	52 ,3	10—160	−0 ,003		I
53. " + Ethylenecyanide	364	103 Timmermans	103 ,3	1—150	−0 ,008		I
54. " + Nitromethane	364	125 Rothmund	118	5—165	−0 ,001		I
55. " + Furfurol	364 —419	67 ,6 Van der Zee	—	1—180	+0 ,004	A Maximum.	IIa
56. " + Phenol	364 —419	69 Rothmund	68 ,9	1—150	+0 ,0033		IIa
57. " + p-Nitrophenol	364	90 ,5 Timmermans	94 ,3	1—120	+0 ,01		IIa
58. " + m-Nitrobenzoic acid	364	107 ,5 Alexejeff	107 ,3	5—125	+0 ,008		IIa
59. " + Benzoic acid	364	115 ,5 Alexejeff	118 ,5	5—200	+0 ,0025		IIa

N.B In salicylic acid + water the observations are impossible, because the acid always crystallises before the upper critical point is reached.

60. Water + Aniline	364°—426°	164°,5	165°,0	10—210	+0°,009		IIa
61. " + Triethylamine L	364 —259	18 ,3 Kuenen	—	1—146	+0 ,02	A Maximum.	III
" + " L	364 —259	19 ,0 Rothmund	18 ,0	1—90	+0 ,02		III
62. " + Collidine L	364 —	6 ,5 Rothmund	1 ,3	1—200	+0 ,0045		III
63. " + Nicotine L	364 —	61 ,0 Hudson	61 ,3	1—200	+0 ,01	A Maximum. By reaction of the nicotine with the glass the upper critical point is not to be reached.	III
64. " + Bromalhydrate L	364 —	—	103 ,4	5—155	+0 ,027	A Maximum. No Maximum.	III
65. Sulphur + Monochlorbenzene	—360	116 Alexejeff	117 ,0	5—85	−0 ,025	The Cailletet apparatus with tube was placed upside down, and instead of mercury water was used as pressure liquid.	I
66. Ternary system: Pyridine + water + natriumcarbonate	A mixture which had a region of non-miscibility between 55° and 65° under three-phase pressure in the critical region, was quite homogeneous at 130 atms.						

(1) Studied by Mr. LECAT in his Thesis for the doctorate, Brussels, 1909.

TABLE II.

System propane + methylalcohol. p,t-projection of the critical line (observations of Kuenen)	t. 21°,15	p. 10 atm.	$\frac{\partial t}{\partial p}$	−0°,12
	20 ,80	13 "		−0 ,075
	20 ,05	23 "		−0 ,07
	19 ,85	26 "		−0 ,053
	19 ,40	34,5 "		−0 ,035
	19 ,00	46 "		−0 ,044
	18 ,60	55 "		−0 037
	18 ,05	70 "		−0 ,027
	17 ,65	85 "		−0 ,030
	17 ,20	100 "		
System isobutane + methylalcohol (observations of Timmermans)	t. 20°,10 ± 0°,05	p. 10 atm.	$\frac{\partial t}{\partial p}$	−0°,0017
	20 05 ± 0 ,05	40 "		+0 ,0017
	20 ,10 ± 0 ,05	70 "		+0 ,007
	20 ,30 ± 0 ,05	100 "		+0 ,009
	20 ,65 ± 0 ,1	140 "		

were always made starting from the homogeneous region towards the heterogeneous region. All the measurements were repeated till the results differed less than the error of observation, which was naturally rather large in these measurements.

In this way results were reached, which were sufficiently accurate for our purpose, as appears from the following comparison of values for the change of the plaitpoint temperature with the pressure found by us and by VAN DER LEE and KUENEN.

	$\frac{dt}{dp}$ according to T. and K.	$\frac{dt}{dp}$ according to VAN DER LEE and KUENEN
Phenol and water	+ 0.0033	+ 0.004
Methyl alcohol and hexane	+ 0.032	+ 0.029
Triethylamine and water	+ 0.02	+ 0.02

We may state in conclusion that all the measurements were carried out by the first mentioned of us (T.).

6. *Results.*

The adjoined table contains a survey of the obtained results. Column I gives the examined system, Column II the critical temperatures of the components for so far as they are known, Column IV the critical end-points (plaitpoint in presence of the third phase); the cases in which the three phases exist at *higher* temperature are indicated by an L (lower critical end-point). In column III the values are found obtained by other investigators. Column V gives the limits of pressure within which the observations were made, column VI the quotient $\frac{dt}{dp}$, which indicates how the plaitpoint temperature varies with an increase of pressure of one atmosphere. Column VII contains special remarks and data about the vapour pressure line of the mixture, for so far as they are known[1]), in connection with the above mentioned theorem of VAN DER WAALS. Column VIII indicates to what type of the classification, which will be treated in the next §, the system belongs. In connection with this some systems of other investigators, which have not been investigated by us, have been inserted in the table.

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[1]) A survey of the known maxima in vapour-pressure lines is found in KUENEN, loc. cit. p. 129 and in SYDNEY YOUNG, Fractional Destillation. MACMILLAN and Co, London 1903 p. 67.

7. *Survey of the possible shapes of the plaitpoint curve.*

We shall give a summary of the possible shapes of plaitpoint lines, for so far as they might be actually found in our investigations, so without considering all the cases to which the theory leads (e.g. for cases as helium and water). This summary has been chiefly derived from the above mentioned investigations of VAN DER WAALS, to which we must refer for further particulars. There also T,x and v,x projections are found of the plaitpoint lines discussed here in p,T projection.

We first consider the case that the region of non-miscibility does not extend to the region of the critical temperatures liquid-vapour. The simplest imaginable case is this that at a certain temperature a small closed plait is formed on the spinodal curve of the transverse plait, which extends, and exceeds the limit of the stable region, remaining closed towards the sides of the small volumes all through according to our supposition mentioned on p. 237. At higher temperature this plait contracts again, it retreats again within the transverse plait, and at last disappears on the spinodal curve of the transverse plait in an heterogeneous double plaitpoint. Or strictly speaking: it disappears on the spinodal curve, since in this case there is only question of *one* spinodal curve. Whether in this case we should speak of a longitudinal plait, or only of a branch plait which forms and disappears again, depends on this whether in the nomenclature we pay special attention to the binodal curve, or to the spinodal one. For the above reason (appearance of the solid state) we cannot as a rule reach experimentally the point where the plait egresses into the stable region, but only that where it retreats again into the metastable one, therefore we call this case that of the "retreat". Fig. 1 represents the p,t-projection of the plaitpoint line for this case schematically [1]). AB is the plaitpoint line vapour-liquid, CD the plaitpoint line of the longitudinal plait with two cusps (heterogeneous double plaitpoints) and a maximum of the pressure, situated at the highest pressure E which the plaitpoint turned towards the small volumes reached after its egress from the transverse plait. So at higher pressure than E non-miscibility cannot be brought about in any way. The line FG indicates the three-phase-pressure; F and G are critical end-points, of which, as we said, generally only G is to be realized.

Of course it is not excluded that F and G draw nearer together,

[1]) In this figure and in figures $2a$ and $2b$ the point A must be taken as lying to the right of D and I.

and at last coincide and disappear, which means that the longitudinal plait does not cross the binodal curve of the transverse plait, but remains entirely hidden inside it (e. g. at negative pressures). Then non-miscibility could only be observed in consequence of retardation phenomena; we have then what in a former paper TIMMERMANS has called a negative saturation curve.

But theory has taught and experiment has confirmed that there exists a second, more complicated type, that of the splitting up of a plait, which is represented in fig. 2. On the part DE of the plaitpoint line two homogeneous double plaitpoints have arisen, in H and I, so that the plaitpoint line, after having descended in temperature from D to H, rises again to I, after which it descends again through E to C. In this case two possibilities may present themselves. Either the three-phase pressure line will run from a point between C and E to a point between I and H, or the upper end-point of the three-phase pressure will lie between H and D. In the former (more probable) case we shall get into the heterogeneous region, if the pressure is raised in the upper end-point, in the second case in the homogeneous region, and increasing the pressure still more at constant temperature, we shall either get into the heterogeneous region or not, according as G lies at higher or lower temperature than I. Whether we pay attention to the binodal or the spinodal line, in any case we shall have to speak here of a separate longitudinal plait. For the further elucidation of the difference between $2a$ and $2b$, we give also the v,x-projection of the binodal and the spinodal curve for this case in fig. $6a$ and $6b$ at the temperature of the homogeneous double plaitpoint.

Let us then consider the case for which the region of non-miscibility extends into the critical region proper. The plaitpoint lines of longitudinal plait and transverse plait, which were separate in the preceding figures, will now unite to the plaitpoint line of one plait, which we may now more appropriately call a "complex" plait. As we said before, we shall not discuss here all the possibilities of a complex plait, but only those which are important for our investigation. The simplest case now of a complex plait is again that only two heterogeneous double plaitpoints occur. Then we shall get fig. 3. The realizable part of the plaitpoint line begins in A, the critical point of the first component, and runs to the critical end-point G in the presence of a second liquid phase. The real non-miscibility of two liquids begins in the lower end-point F, which may, of course, lie considerably below A, but the coexistence between two liquids, as it would in the beginning undoubtedly be called in this case,

passes gradually into a coexistence vapour-liquid, till B, the critical point of the second component, is reached.

If now in this case splitting up of a plait takes place, we might have the case of fig. 4, analogous to fig. 2. Here, too, we might distinguish two sub cases 4a and 4b, analogous to 2a and 2b. And also a case like fig. 5, would be possible.

It is not easy to state in a single word in what relation the classification adopted here stands to that of BÜCHNER and VAN LAAR, because in our opinion by the latter cases have been classed together which should be sharply distinguished, and on the other hand what belongs together, has been separated. For BÜCHNER's type II, VAN LAAR's type III [1]) comprises the cases of retreat and splitting up, which are entirely different both theoretically and experimentally. BÜCHNER's fig. 10, VAN LAAR's figs. 11 and 2a may therefore be found in our figs. 1, 2a and 2b. On the other hand BÜCHNER's types I and III, VAN LAAR's I and II (first paper) are distinguished only in this, that there are one maximum and one minimum of the pressure more in the last plaitpoint line, a circumstance, which is not of great importance physically, as little as the occurrence of a maximum or a minimum of the pressure in the ordinary plaitpoint line is generally considered to be of importance (in opposition to a temperature minimum or maximum). We find these two types, therefore, combined in our fig. 3, if we consider that F may lie considerably below A, and bear in mind what has been said above about the assumption of open plaits. There would be an essential difference between fig. 6 and 13 of BÜCHNER, fig. 1 and 7 of VAN LAAR's first paper only when the line KPD (fig. 13 loc. cit.) was imagined to show a temperature minimum and maximum in its further course; then fig. 13 resp. 7 would agree with fig. 5 here. Neither BÜCHNER nor VAN LAAR, however, mentions this.

In VAN LAAR's later paper type II has been modified and has become different from I in so far, that now not one pair of heterogeneous double plaitpoints occur, but two pairs (resp. one pair and an open plait, which comes to the same thing). Such a complicated system of plaits cannot be arrived at on the simple suppositions of VAN LAAR,

1) It is remarkable that VAN LAAR, who on p. 669 of his first paper decidedly notices the difference in the systems with positive and with negative dp/dt, does not say a word about it in his later paper. In connection with this the dotted line of fig. 11 of the former paper, which refers to positive dp/dt has been left out in the figure of the later one, probably because the special suppositions of VAN LAAR render the occurrence of the required homogeneous double plaitpoints impossible for normal substances.

who ascribes, therefore, the appearance of this type to abnormality. The occurrence of two heterogeneous double plaitpoints R_1 and R'_2 by the side of the open plait $C_0 R_2 A$ is, indeed, not very probable, nor do any theoretical or experimental grounds plead in favour of the existence of such a system of plaits. That, however, VAN LAAR's original type II, BÜCHNER's type I, fig. 3 here, is very well possible for normal substances, VAN DER WAALS's theoretical investigations and BÜCHNER's experimental ones have proved convincingly.

8. *Classification of the investigated systems.*

All the systems mentioned in the table appear to belong to the types 1, 2, or 3, except one single system, methyl alcohol and ethane, which may possibly belong to type 5.

For normal components we have found type I for nitro-benzene and aniline with saturated hydro-carbons, a few times also the case of fig. 2*a* (SO_2 with hexane); the cases of CO_2 with little volatile substances, chrysene and ether, etc. found by BÜCHNER, belong to type 3.

We have not met with the case 2*b* for normal substances, nor can we state with certainty that we have found it for abnormal substances, for on account of the very high pressures which soon occur, the plaitpoint line cannot be examined far enough. But if we compare inter se the results which a series of homologous hydro-carbons with methyl alcohol give, the supposition suggests itself that we have to do here with a transition of case 2*b* to 2*a*, passing from the lower to the higher hydro-carbons.

For while for propane and methyl alcohol according to the observations of KUENEN the $\frac{dt}{dp}$ is $-0°,12$ starting from the critical end-point (so we have here case 1 or 2*b*), this value changes to $-0°,03$ at about 100 Atm., which certainly agrees better with 2*b*. (See table II).

For isobutane [1]) and methyl alcohol the $\frac{dt}{dp}$ is at first practically speaking 0; at 10 or at 70 atms. we find no perceptible difference for the plaitpoint temperature; if the pressure is increased further, the temperature too begins to rise very perceptibly, and $+0.009$ is found for $\frac{dt}{dp}$ at 140 atm. (See table II). So we are forced to conclude

[1]) The preparations of butane and isobutane were particularly pure; they had been presented to us by Mr. OUEDINOFF at Brussels, for which we once more express our thanks to him.

that for this system the points G and H nearly coincide, or in other words that in fig. 6 the line AB passes about through the double point.

Butane and methyl alcohol then at once begin with a rise of temperature with increasing pressure, though it is very small (+0.007) at first, and the successive homologues begin then with always higher positive value of $\frac{dt}{dp}$: Isopentane with + 0.02, hexane with 0,03, decane with 0.04.

In connection with this the question rises if really the first system of this series, methyl alcohol with ethane investigated by KUENEN, is simply a case of 3; or if it should be considered as an example of type 5, which is analogous to 2, a question which we shall not try to decide for want of data.

We have not been able to reach the point E in any of our binary systems [1]), here too, probably on account of the rapid increase of the pressure. Some of the systems of water with bases of the pyridine-series are examples of this type, which present a perfectly closed plait, for which therefore in any case such a point must occur, and these plaits being of small extension, we may expect to succeed in reaching the point E. We hope shortly to be able to take this investigation in hand.

We may remark in conclusion that in all the cases, in which the vapour-pressure line of the system has been determined, the rule derived by VAN DER WAALS, appears to hold: Splitting up of a plait takes place only when there is a maximum in the vapour-pressure line (min. critical temperature of the unsplit mixture).

Amsterdam. Physical Laboratory of the University.

Chemistry. — "*On the system water-natrium sulphate*". By Prof. A. SMITS and Dr. J. P. WUITE. (Communicated by Prof. A. F. HOLLEMAN).

Already before [2]) one of us succeeded in showing that the system *water-natrium sulphate* belongs to the type *ether-anthraquinone*, in which the probability was also pointed out that this would also be the case for the other systems water-salt which show a retrogressive solubility curve.

As the above behaviour was found in a preliminary investigation,

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[1]) We have reached it, however, in the ternary system: Pyridine + water + natrium carbonate.

[2]) SMITS. Inaugural address. Amsterdam, Dec. 9, 1907, p. 19.

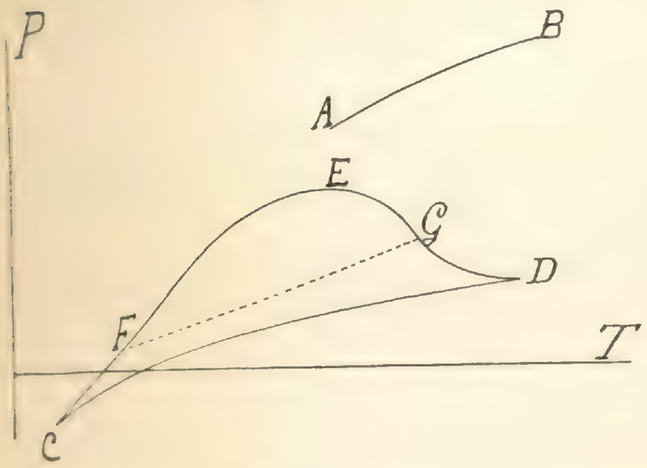

Fig. 1.

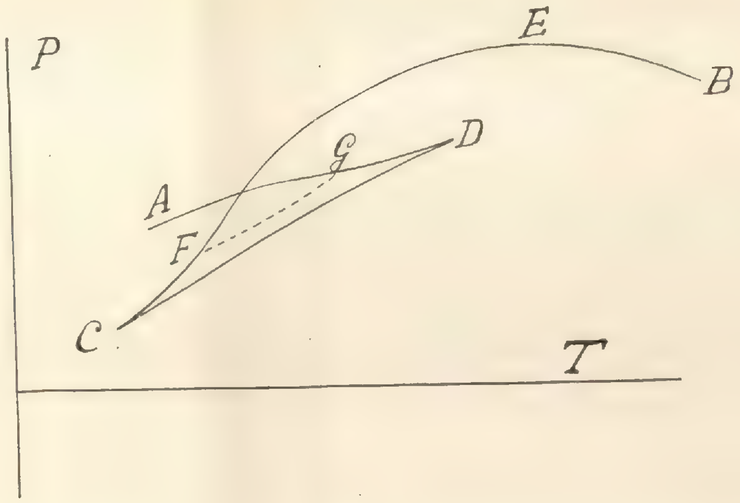

Fig. 3.

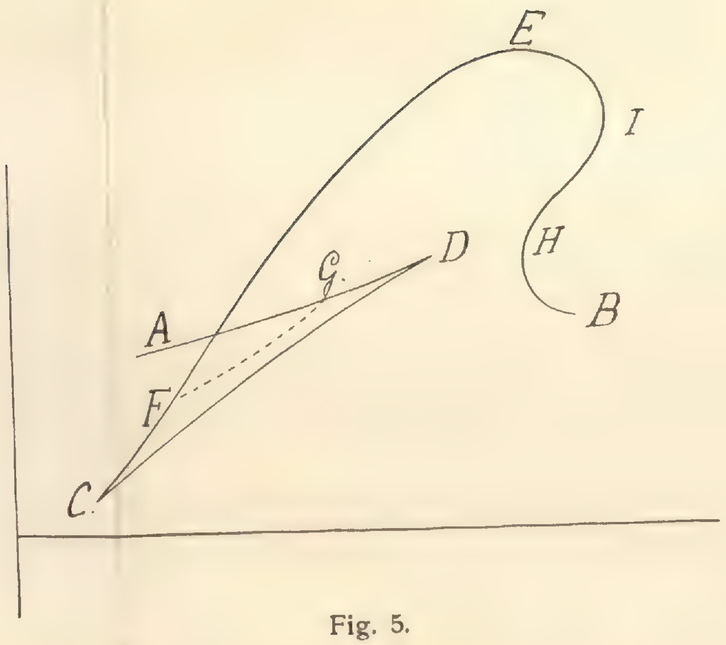

Fig. 5.

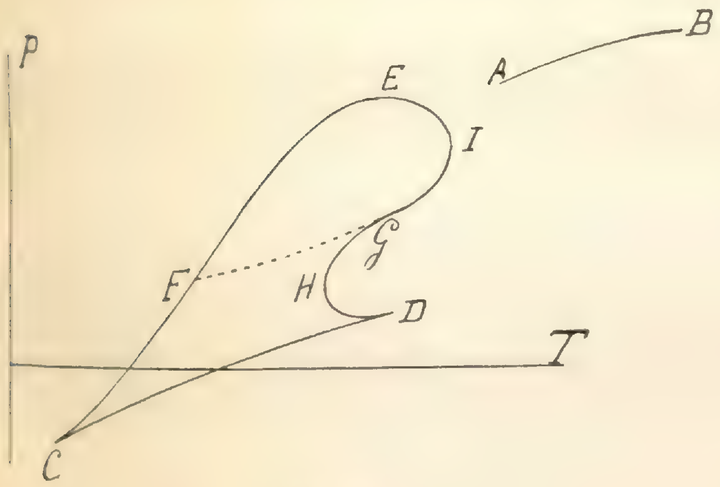

Fig. 2*a*.

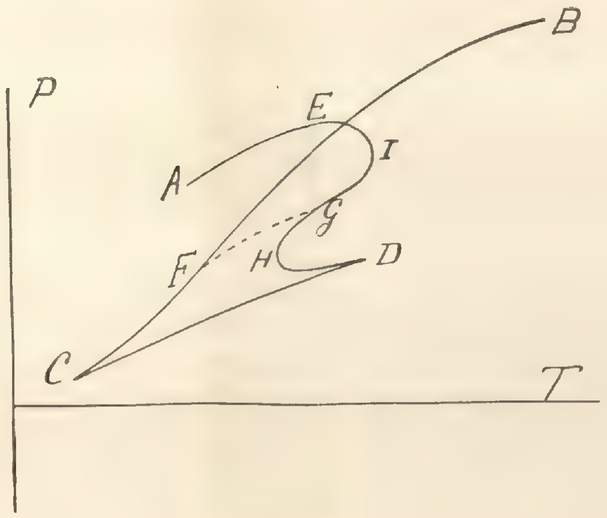

Fig. 4*a*.

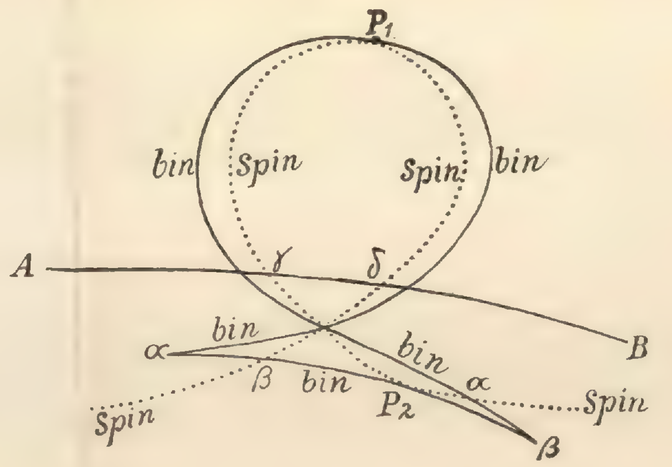

Fig. 6*a*.

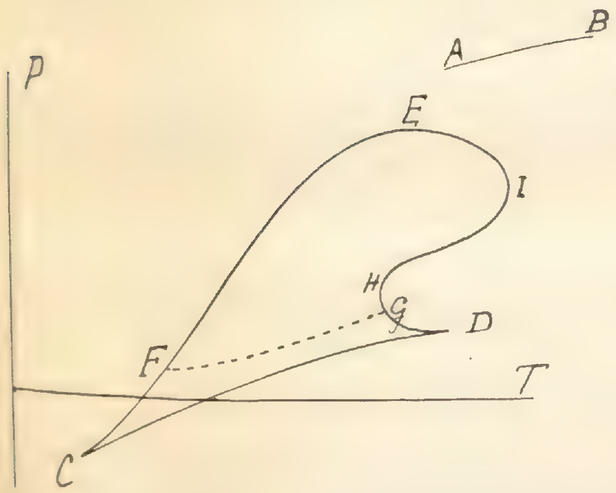

Fig. 2*b*.

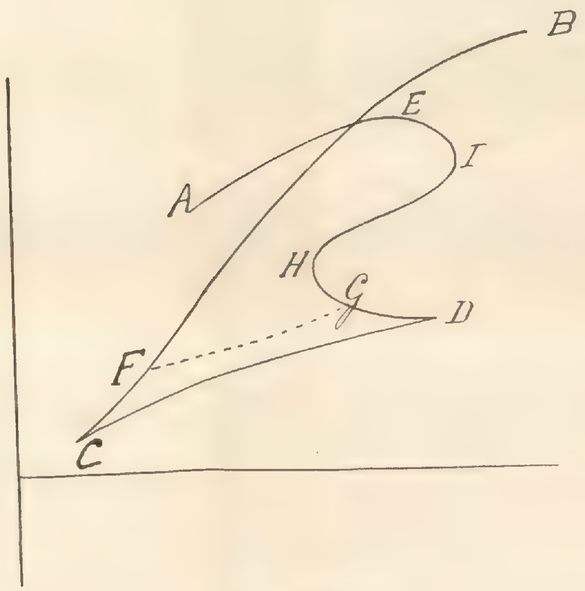

Fig. 4*b*.

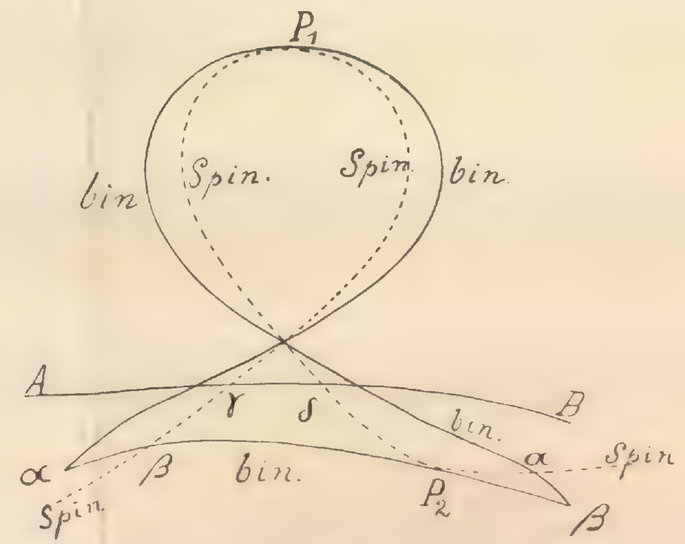

Fig. 6*b*.

which was almost exclusively restricted to the critical neighbourhood, and the system water-natrium sulphate presents so many remarkable peculiarities also outside this critical region, it was resolved to take in hand an extensive quantitative investigation of this system, the results of which will briefly be communicated here.

The first part of this investigation dealt with the determination of the vapour pressure lines.

Fig. 1 represents what was found schematically, the temperature and the pressure being accurately indicated at the most important points.

One part of this figure has been reproduced in Fig. 2 on a larger scale in the real proportions and contains among others the three-phase-curves for

$Na_2SO_4 . 10\,aq + L + G$, $Na_2SO_4 + L + G$, $Na_2SO_4 . 10\,aq + Na_2SO_4 + G$, $Na_2SO_4 . 7\,aq + L + G$, $Na_2SO_4 . 7\,aq + Na_2SO_4 + G$,, which have all been measured accurately to 0,1 mm. by means of a very simple apparatus with mercury manometer.

From the situation of these lines, which had already been indicated schematically by VAN 'T HOFF,[1]) the situation of two quadruple points *E* and *D* follows with pretty great certainty, which is perhaps best proved by this that the temperature of the point *E* agrees perfectly with the exceedingly accurate determination of the transition temperature by RICHARDS and WELLS [2]), who found 32.384° for the transition point of the transformation $Na_2SO_4 . 10\,aq \rightleftarrows Na_2SO_4 + aq$ under a pressure of 1 atm., which differs very little from that under the vapour tension pressure *E*. (See table p. 247).

It follows in the first place from these tables that the three-phase curve for $Na_2SO_4 + L + G$ lies between the two others, which the theory requires, and further that for **32.4°** the three three-phase pressures have the same value, viz.: **30.8** mm. Hg., from which follows that at this temperature and pressure $Na_2SO_4 . 10\,HO$, Na_2SO_4, L and G are in equilibrium with each other, which may be considered as in perfect harmony with RICHARDS and WELLS' observation.

If we now apply VAN 'T HOFF's formula to the table for the three-phase curve $Na_2SO_4 . 10\,aq + Na_2SO_4 + G$ in the same form as was used by FROWEIN [3]), we get:

$$Q = 4.606\,\frac{T_1 - T_2}{T_1\,T_2}\left(log\,\frac{P'_2}{P_2} - log\,\frac{P'_1}{P_1}\right),$$

[1]) Vorlesungen I, 58.
[2]) Zeitschr. f. physik Chem. **43**, 471, (1903).
[3]) Z. Ph. Chem. I, 8 (1887).

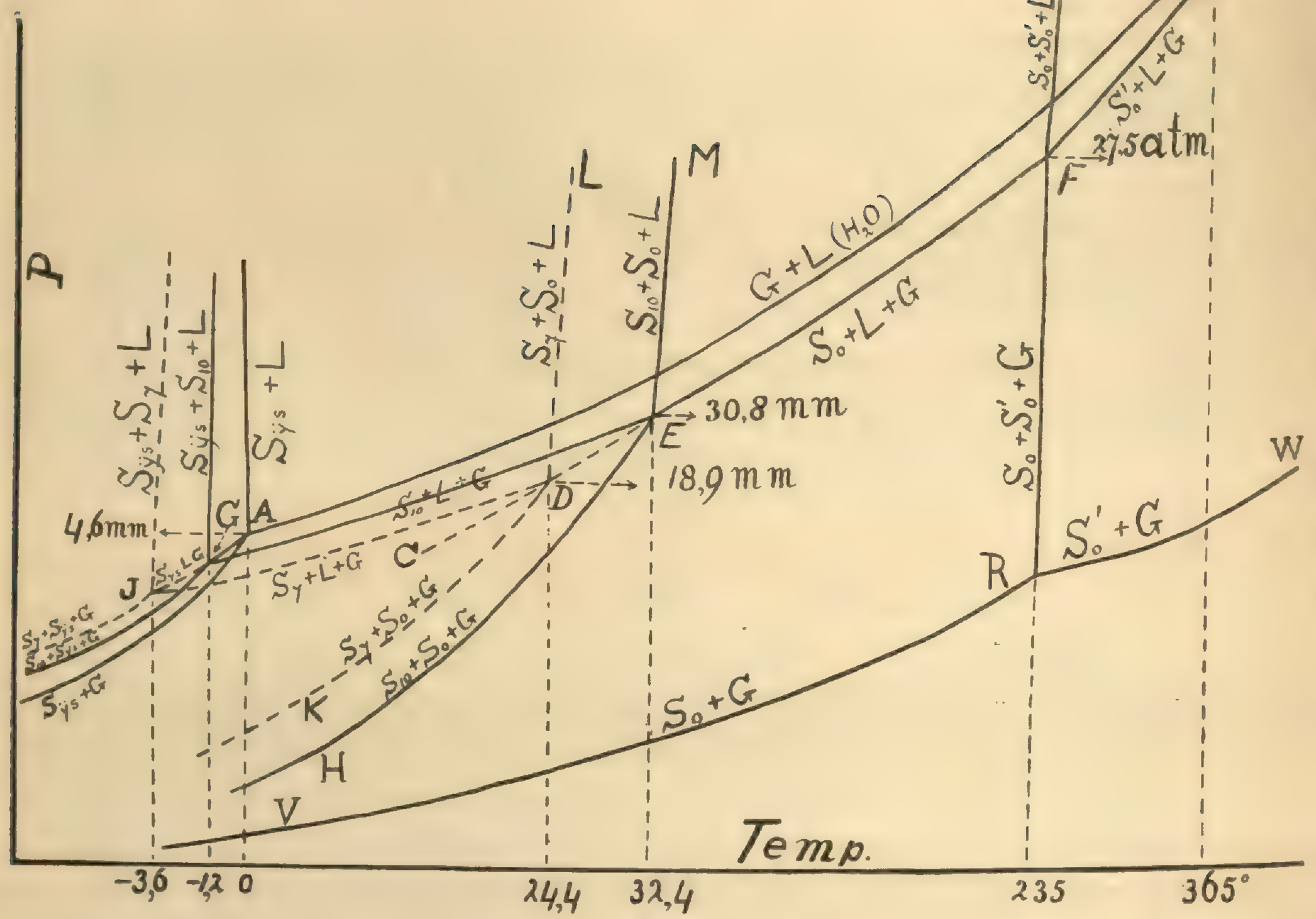
P
200atm
B
p
L
M
$S_{11}+S_0+L$
$S_{11}+S_{10}+L$
$S_{11}+S_0+L$
$S_{10}+S_0+L$
$G+L(H_2O)$
S_0+L+G
$S_0+S_0'+L$
$S_0'+L+G$
27,5atm
F
30,8 mm
E
18,9 mm
D
4,6mm
G
A
$S_{10}+L+G$
$S_\gamma+L+G$
C
J
$S_\gamma+L+G$
$S_\gamma+S_0+G$
$S_{10}+S_0+G$
K
H
V
$S_{11}+S_\gamma+G$
$S_{11}+S_{10}+G$
$S_\gamma+L+G$
$S_{11}+S_\gamma+L$
$S_{11}+S_{10}+L$
$S_\gamma+L$
$S_{11}+S_\gamma+S_0+L$
$S_{11}+S_0+L$
$S_{10}+S_0+L$
S_0+L+G
S_0+G
$S_0+S_0'+G$
R
$S_0'+G$
W
Temp.
-3,6
-1,2
0
14,4
32,4
235
365°

Fig. 1.

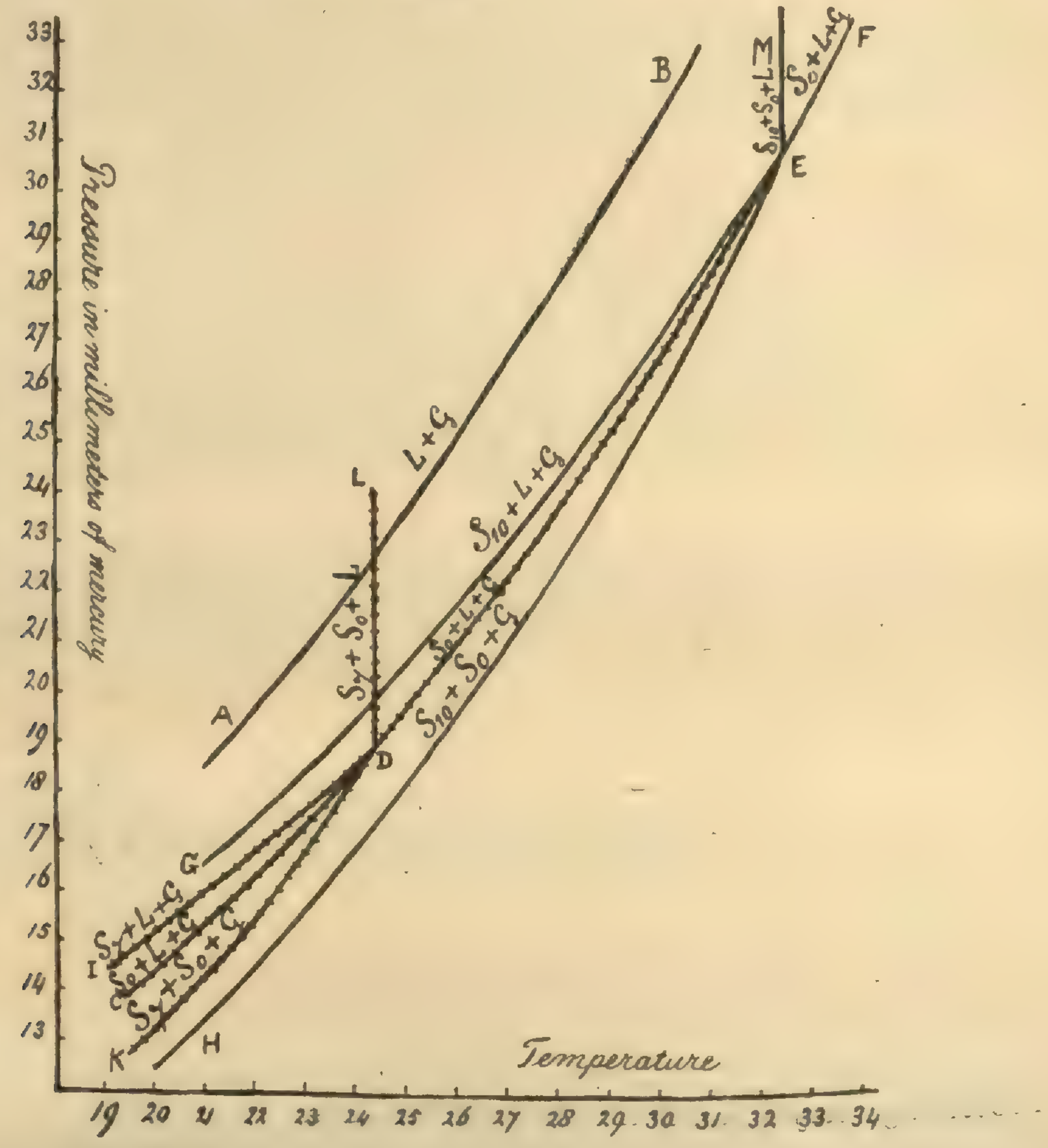
Pressure in millimetres of mercury
33
32
31
30
29
28
27
26
25
24
23
22
21
20
19
18
17
16
15
14
13
B
F
E
$S_{11}+S_0+L+G$
S_0+L+G
L+G
$S_{10}+L+G$
$S_\gamma+L+G$
S_0+L+G
$S_{10}+S_0+G$
A
D
G
$S_\gamma+L+G$
I
$S_\gamma+L+G$
C
$S_\gamma+S_0+G$
K
H
Temperature
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34

Fig. 2.

For the first mentioned three-phase lines was found:

Three-phase line $Na_2SO_4\,10\,aq. + L + G$		Three-phase line $Na_2SO_4 + L + G$		Three-phase line $Na_2SO_4\,10aq + Na_2SO_4 + G$	
Temp.	Pressure in m.m. Hg.	Temp.	Pressure in m.m. Hg.	Temp.	Pressure in m.m. Hg.
21	16.4	19.4	13.9	20.2	12.5
22.4	17.8	19.8	14.2	21.6	14.1
22.6	18.1	20.6	14.9	23.0	15.7
23.0	18.5	21.6	15.9	24.1	17.0
23.2	18.7	22.6	17.0	25.0	18.1
23.6	19.2	23.5	18.0	25.9	19.4
25.0	20.6	24.6	19.2	27.0	21.0
26.1	21.8	25.8	20.6	28.1	22.7
27.0	23.0	28.1	23.9	29.5	25.0
28.2	24.7	30.5	27.6	31.0	27.9
29.4	26.3	**32.4**	**30.8**	**32.4**	**30.8**
29.9	27.0	32.8	31.4		
31.0	28.6	33.8	33.5		
32.4	**30.8**	35.0	36.0		
		37.0	**40.1**		
		39.0	44.6		
		40.4	48.2		
		41.7	52.3		
		44.7	61.9		
		48.6	75.5		

in which P_1 and P_2 are two three-phase pressures at the absolute temperatures T_1, T_2, P'_1, and P'_2 denoting the vapour tension of pure water at the same temperatures.

Q indicates then the heat-effect in the formation of decahydrate from 1 gr. mol. of liquid water and anhydride.

If we take the pressures corresponding with 20° and 32°4, we find for $Q = 2440$ cal., from which follows that the total heat of hydration of $Na_2\,SO_4\,.\,10\,H_2O = 24400$ cal.

We may mention here at the same time that this indirect deter-

mination of the heat of hydration appeared to be the only accurate one, as a direct measurement of the heat effect was not possible on account of the slow reaction.

The point D is a metastable quadruple point, where in metastable state the phases $Na_2SO_4 . 7\,aq$, Na_2SO_4, L and G are in equilibrium. For this transition point (under a pressure of 1 atmosphere), where the transformation.

$$Na_2SO_4 . 7\,aq \rightleftarrows Na_2SO_4 + aq$$

takes place, we find graphically from the solubility curves of LOEWEL and GAY-LUSSAC the temperature **24,4°**.

As may be derived from the subjoined table, a very pronounced break was found at 24°,4 in the determination of the three-phase line for $Na_2SO_4\,7\,aq + Na_2SO_4 + G$, from which follows that the transition point under the vapour pressure also lies at 24°,4.

Three-phase curve.

$Na_2SO_4 . 7\,aq + Na_2SO_4 + G$

Temp.	Pressure in m.m.Hg.	Solid phases
20.0	13.2	$7\,aq + anh + G$
21.0	14.3	„
22.0	15.4	„
22.9	16.6	„
23.6	17.6	„
23.9	18 0	„
24.1	18.3	„
24 4	**18.9**	$7aq + anh + L + G$
24.6	19.2	$anh + L + G$
24.9	19.6	„
25.3	20.0	„

If prolonged to this temperature, the three-phase curve for $Na_2SO_4 . 7\,aq + L + G$ might have given a confirmation, but for this purpose we ought to have taken exactly the concentration corresponding to the transition point, 6,15 mol $^0/_0$ Na_2SO_4, which would be nearly always exceeded on account of particular experimental difficulties, in consequence of which by the side of the hydrate with 7 aq the anhydride would always appear, and so the

three-phase equilibrium $Na_2SO_4\ 7\ aq + Na_2SO_4 + G$ would always be measured.

This is, therefore, the reason that the three-phase line $Na_2SO_4\ 7\ aq + L + G$ could only be continued to 23° as appears from the following table.

Three-phase curve.

$Na_2SO_4 . 7\ aq + L + G$

Temp.	Pressure in m.m.Hg.
19.1	14.5
20.0	15.3
21.0	16.1
21.9	16.9
23.0	17.9

Before leaving fig. 2, we mention that the transition line DL has not been examined, the transition line EM, however, has, viz. by TAMMANN [1]), who found when pursuing the investigation up to high pressures, that this transition curve has a vertical tangent, which appears clearly from the following values.

Three-phase pressure.

$Na_2SO_4\ 10\ aq + Na_2SO_4 + L$

Temp.	Pressure in $\frac{K.\ Gr.}{cm^2}$
32.6 + 0.045	100
+ 0.079	200
+ 0.122	460
− 0.043	1000
− 1.19	2000
− 3.43	3000

We may finally remark that AB denotes the vapour pressure curve of pure water, and VRW the sublimation curve of the two modifications of Na_2SO_4.

If we now return to fig. 1, it is noteworthy, that the three-phase curve for $Na_2SO_4 + L + G$ had already been determined by TAM-

[1]) Kristallisieren und Schmelzen, 254.

MANN [1]) from 50° to 100°; these determinations, however, were repeated before we proceeded to higher temperature, that the observations might be perfectly comparable.

Before proceeding to mention the results, we state that the observations to the temperature of 50° were carried out in an apparatus with mercury manometer, which had been exhausted with particular

Three-phase curve.

$Na_3SO_4 + L + G$

Temp.	Pressure in m.m.Hg.
50.6	83.6
53.8	99.0
56.8	114.0
59.6	130.0
62.8	151.0
65.6	171.0
69.4	200
69.8	208
74.8	258
80.2	321
86.0	403
90.6	482
95.2	574
100.0	681
105.0	812
111.0	1009
116.1	1196
119.8	1295
124 8	1569
125.8	1617
129.8	1833
135.6	2172
140.8	2521

[1]) Wied. Ann. **24**, 530 (1885).

care, and was entirely immerged in a thermostat. From 50° to 140° a slightly different method was followed; the mercury manometer in the thermostat was used as an indicator, as is usually done, while the pressure was read on an open manometer, which was connected with the small manometer by means of a *T*-joint with cock, and was placed outside the bath, the pressure being kept equal to the three-phase pressure (by means of a cycle-pump).

The investigation from $\pm 50°$ to $\pm 140°$ gave the following result. (See table p. 250).

These observations, which harmonize very well with those of TAMMANN up to 100°, show that this three-phase curve does not present any irregularity up to 140°.

As pressures above 3 atmospheres were dangerous for the apparatus which was used up to 140°, we now adopted the method with the Cailletet tube, which was up to 217° of glass of a high melting point, and above this temperature of quartz, because glass is rapidly attacked by water above 200°.

Unfortunately, however, quartz was not the right material for the observation at high temperatures either; for though quartz was not changed at all by water, even at the critical temperature, it *was* attacked by a solution of Na_2SO_4 under for the rest similar circumstances [probably in consequence of hydrolysis of the salt], which caused the quartz tubes to burst repeatedly, and which rendered the continuation of the vapour tension determinations, which require a comparatively long time, impossible above 260°.

From 140° to 260° the investigation gave the following result. (See table p. 252).

If we compare these pressures with those of pure water at the same temperature, we notice in the first place that as was to be expected in connection with the slight solubility of the natrium-sulphate, the differences are small, and in the second place it strikes us that these differences reach a maximum value at $\pm 234°$, which points to a point of transition, which has been really found by HÜTTNER and TAMMANN [1]) at 235°, and by BOEKE [2]) at 239°. Shortly after NACKEN [3]) showed in an accurate investigation that the results of HÜTTNER and TAMMANN are nearest to the truth, as he found 234° for the point of transition. His experiments revealed further that the modification of Na_2SO_4 is rhombic below 234°, and monoclinic above it.

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[1]) Zeitschrift. An. Chem. **43**, 215.
[2]) Thesis for the doctorate, Amsterdam 1906.
[3]) Neues Jahrb. für Mineralogie, Beilageband **24**, 27 (1907).

Three-phase curve.
$Na_2SO_4 + L + G$

Temp.	Pressure in atmospheres
141.9	3.5
142.2	3.6
142.4	3.7
144.7	3.9
145.0	3.9
147.2	4.1
148.2	4.2
149.0	4.2
151.6	4.5
151.7	4.6
152.0	4.7
153.2	4.9
154.2	5.0
157.0	5.3
160.3	5.7
162.5	6.1
163.4	6.2
166.9	6.8
170.4	7.3
172.4	7.7
174.4	8.1
177.3	8.6
179.4	9.2
181 4	9.6
184.1	10.3
186.1	10.7
187.6	11.0
192.6	12.3
201.3	14.2
206.4	16.5
210.2	18.2
217.2	20,4

Three-phase curve.
$Na_2SO_4 + L + G$

Temp.	Pressure in atmospheres	Difference with the vapour tension of pure water.
220	21.2	1.6
222.8	22.2	2.0
226.4	23.9	1.9
230.8	25.6	2.3
233.6	27.1	2.3
234.0	27.1	2.3
237.2	28.9	2.2
240.3	31.0	2.0
244.7	33.7	2.0
247	35.2	1.9
250.4	38.5	1.8
255.2	41.4	1.6
260	45	

The second part of the investigation consisted of the determination of the melting-point line under the vapour pressure, of which fig. 3 gives the examined region.

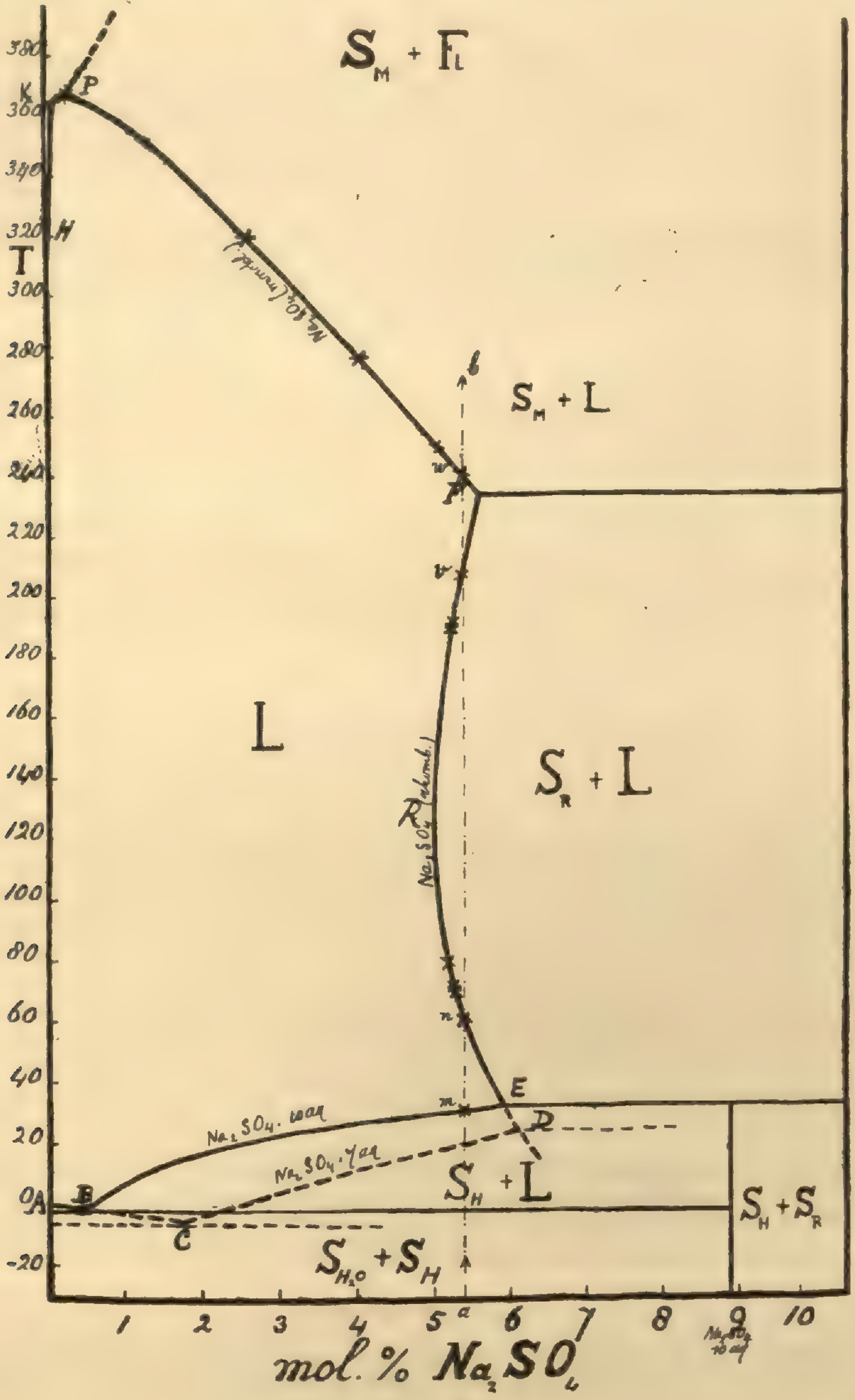

Fig. 3.

The ice-line ABC has already long been known from the investigations of RAOULT [1]), LOOMIS [2]), DE COPPET [3]), and LOEWEL [4]), while

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[1]) Zeitschr. f. physik. Chem. **2**, 489 (1888).

[2]) Wied. Ann. **57**, 503 (1896).

[3]) Ann. de Chim. et phys. [4] **25**, 539 (1872) and Zeitschr. f. physik. Chem. **22**, 239.

[4]) Ann. de Chim et phys. [3] **49**, 50 (1857).

also the melting-point line of $Na_2SO_4 . 10 H_2O$, BE and that of $Na_2SO_4 . 7 H_2O$, CD could be derived already before from the determinations of GAY-LUSSAC [1]), LOEWEL, DE COPPET, and BERKELEY [2])..

Further GAY-LUSSAC and BERKELEY have also made investigations on the melting-point line of the anhydride, and pursuing their determinations up to $\pm 100°$, they could already state that we meet here with the special case of a retrogressive melting-point line.

The first observations above 100° were carried out by TILDEN and SHENSTONE [3]) from 100° to 230°, and by ETARD [4]) from 150 to 320°. Their results have been joined in the following tables.

TILDEN and SHENSTONE.

Temp.	Conc. sat. solution in mol. % Na_2SO_4
100°	5.09
120°	5.035
140°	5.05
160°	5.15
180°	5.305
230°	5.55

ÉTARD.

Temp.	Conc. sat. solution in mol. % Na_2SO_4
150°	5.07
190°	5.13
240°	5.15
320°	2.66

TILDEN and SHENSTONE's table, which is sufficiently accurate to justify us in deriving conclusions from it, shows that the melting-point line possesses a vertical tangent between 100° and 140°.

Only one of ÉTARD's determinations has been serviceable, viz. that at 320°, for this led us to suspect that the plaitpoint curve would meet the melting-point curve, which, as was mentioned in the beginning, appeared really to be the case.

In the accurate determination of the melting-point line from 62° to 365° (according to the method of small tubes) [5]) we succeeded in drawing up the most interesting part of the T-x-figure with perfect certainty, and in demonstrating that the system $H_2O - Na_2SO_4$

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[1]) Ann. de Chim, et phys. **11**, 312 (1819).
[2]) Trans. Roy. Soc. 203 A. 209 (1904),
[3]) Trans. Roy. Soc. **175**, 23 (1884).
[4]) C. R. **106**, 206 (1888).
„ „ **113**, 854 (1891).
[5]) Above 250° quartz tubes were used.

belongs to the type ether-anthraquinone with this peculiarity that in this system two transition points occur, and the phenomenon of retrograde solidification along the melting-point line is not found in the critical neighbourhood, but between the two points of transition.

The result of the investigation between 62° and 365° is viz. as follows.

Temp.	Conc. sat. solution in mol. $^0/_0$ Na_2SO_4
62°	5.39
70°	5.27
72°	5.255
80°	5.18
190°	5.255
192°	5.27
208°	5.39
241°	5.39
250°	5.04
279°	4.12
319°	2.56
365°	very small (critical phenomenon)

If we now represent these observations in the T, x-figure, we get the lines ERF and FP with a transition point in F. For clearness' sake the critical point has been drawn at a too great concentration, because else we could not show that in P the melting-point line passes continuously into the vapour-line PH, which has been assumed as coinciding with the axis for H_2O below 320°.

The retrograde character of the melting-point line of rhombic and monoclinic Na_2SO_4 now brings about very peculiar phenomena for concentrations lying between those of the points R and F.

If we e.g. start from a concentration $a = 5.39$ mol. $^0/_0$ Na_2SO_4, we are at the ordinary temperature in the region for Na_2SO_4 10 aq+L; if we raise the temperature, all the solid substance dissolves at 31° in point m, and we get into the region for L.

At 62 , however, rhombic anhydrical Na_2SO_4 deposits in n from

the solution, which deposition at first increases, viz. to $\pm$ 125°. Above this temperature the deposited quantity of solid substance dissolves more and more, and vanishes again entirely at 208° in *v*. We have then got into the liquid region for the second time, and so have observed the phenomenon of retrograde solidification.

If we now continue heating, another change takes place, and *monoclinic* anhydric Na_2SO_4 deposits from the solution at 241° in the point *w*.

The series of observable phenomena is not exhausted yet, the most interesting one is still coming, for if we raise the temperature still more, we see the critical phenomenon appear at 365°, in consequence of the liquid becoming identical with the vapour in the presence of solid Na_2SO_4.

We must, however, point out here that this latter phenomenon requires another volume than the preceding ones; for it is necessary for the points *m*, *n*, *v*, and *w*, that the quantity of vapour is very small, because we want the known total concentration to be equal in these points to the concentration of the liquid, the three-phase pressure yet prevailing.

For the critical phenomenon, however, the volume of the liquid must be equal to that of the vapour at the critical temperature, and so it is clear that for the observation of the critical phenomenon a new filling is required.

As it appeared in this determination that the critical temperature of the saturated solution did not lie sensibly higher than that which was observed for pure water, viz. 365° [1]); we concluded from this that the solubility of Na_2SO_4 in water at the critical temperature is very slight.

Now the question naturally rises what we get at higher temperature. If decomposition were excluded, a point *q* would have to appear below the melting point of Na_2SO_4, so below $\pm$ 885°, where the same liquid as for *p* or in case of non-miscibility another, reached its critical point, and we should then get a *T*, *x*-figure which would closely resemble that for ether-anthraquinone. But as it is highly

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[1]) For the critical temperature of water other investigators found the following:

NADEJDINE 358,1°

BATELLI 364,3°

CAILLETET AND COLARDEAU 200,5°

KOHNSTAMM AND KEESING 364—365°

SMITS 365°

probable that decomposition occurs at higher temperature, we must give up the study of the remaining part of the T,x-figure.

The salts which with water as second component will most likely behave as Na_2SO_4 in connection with their retrogressive melting-point line are the sulphates of *Ca, Cd, Ce, Fe, Gd, La, Mg, Mn, Nd, Ni, Pr, Sr, Th, Yb* and *Zn*, and also the salts Li_2CO_3, Na_2SeO_4, Na_2HPO_4 $CaCrO_4$.

It is highly probable that the same peculiar behaviour will also often be found when we choose another solvent instead of water, in which it might be possible to extend the investigation over the whole of the region of concentration for salts with low melting points on account of the absence of secondary chemical actions.

Anorg. Chem. Laboratory of the University.

Amsterdam, Sept. 1909.

Chemistry. — *"On heterogeneous equilibria of dissociating compounds"*. By Dr. F. E. C. Scheffer. Communicated by Prof. A. F. Holleman.

On Dec. 30th 1905 Dr. A. Smits published a communication on intersections of the plaitpoint curve and the three-phase curve of a dissociating binary compound in these Proceedings. In this paper some of the possible cases were discussed, the phenomena described which may be expected at such an intersection, and a further systematic discussion of the remaining possibilities announced. Prof. Smits, however, ceded the continuation to Prof. Roozeboom, who proposed it to me as a subject for my dissertation; I have only been able to profit by his guidance for a short time. I gladly avail myself of this opportunity to express my thanks to Prof. Smits for the assistance which he has given me in this investigation.

The systems, for which intersection of the plaitpoint curve and the three-phase line of a dissociating binary compound occurs, may be arranged under the six following types.

I. Both the critical end-points lie on the branch AS; see fig. 1. This case was already discussed in the paper mentioned and illustrated by a combined PT- and xT-projection.

II. One critical end-point lies on the branch AS, the second on the branch SM, so between melting point and max. sublimation point (points p_1 and q_2 from fig. 1). From an examination of fig. 2, which represents this case schematically, it will be clear, that the

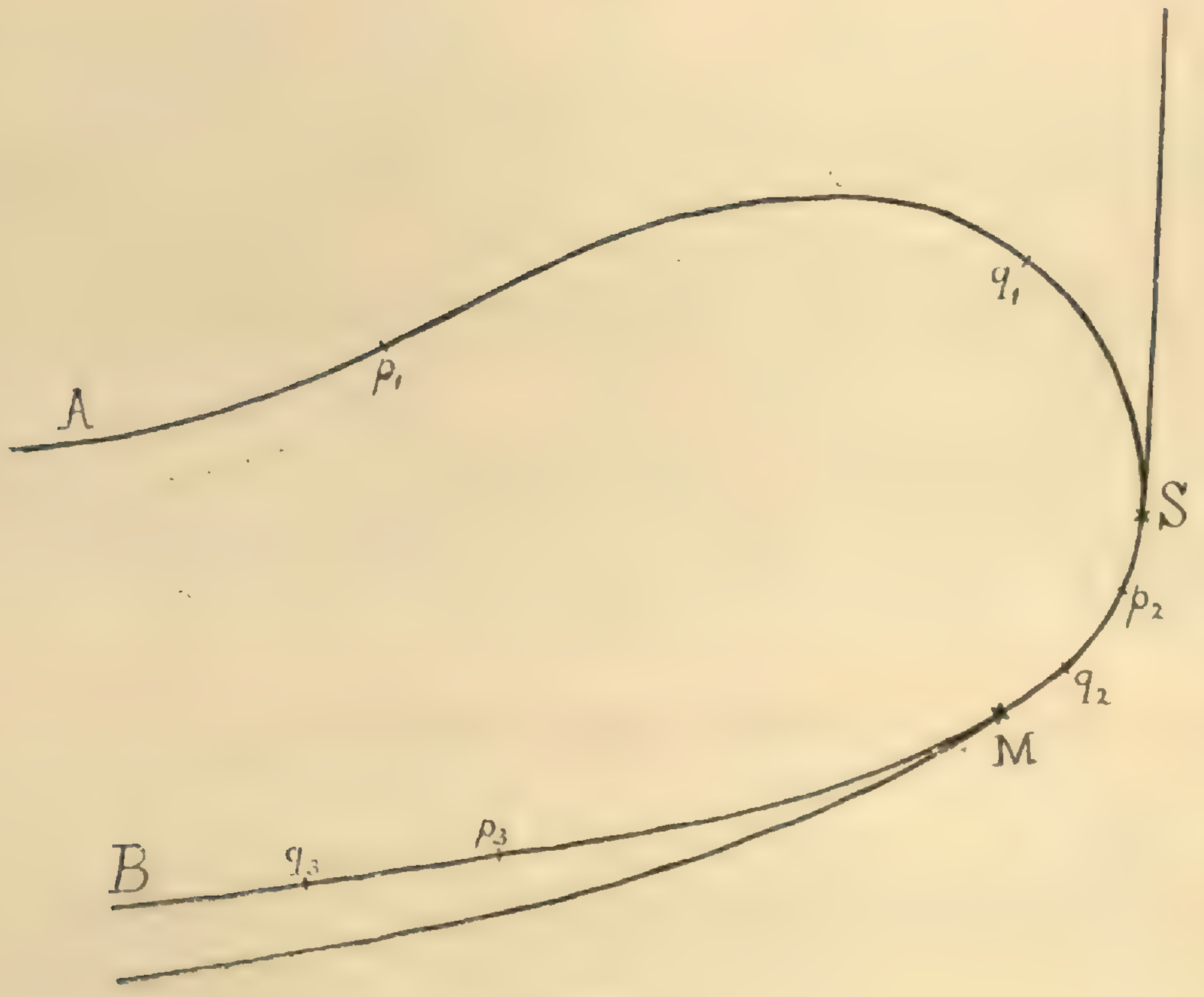

Fig. 1.

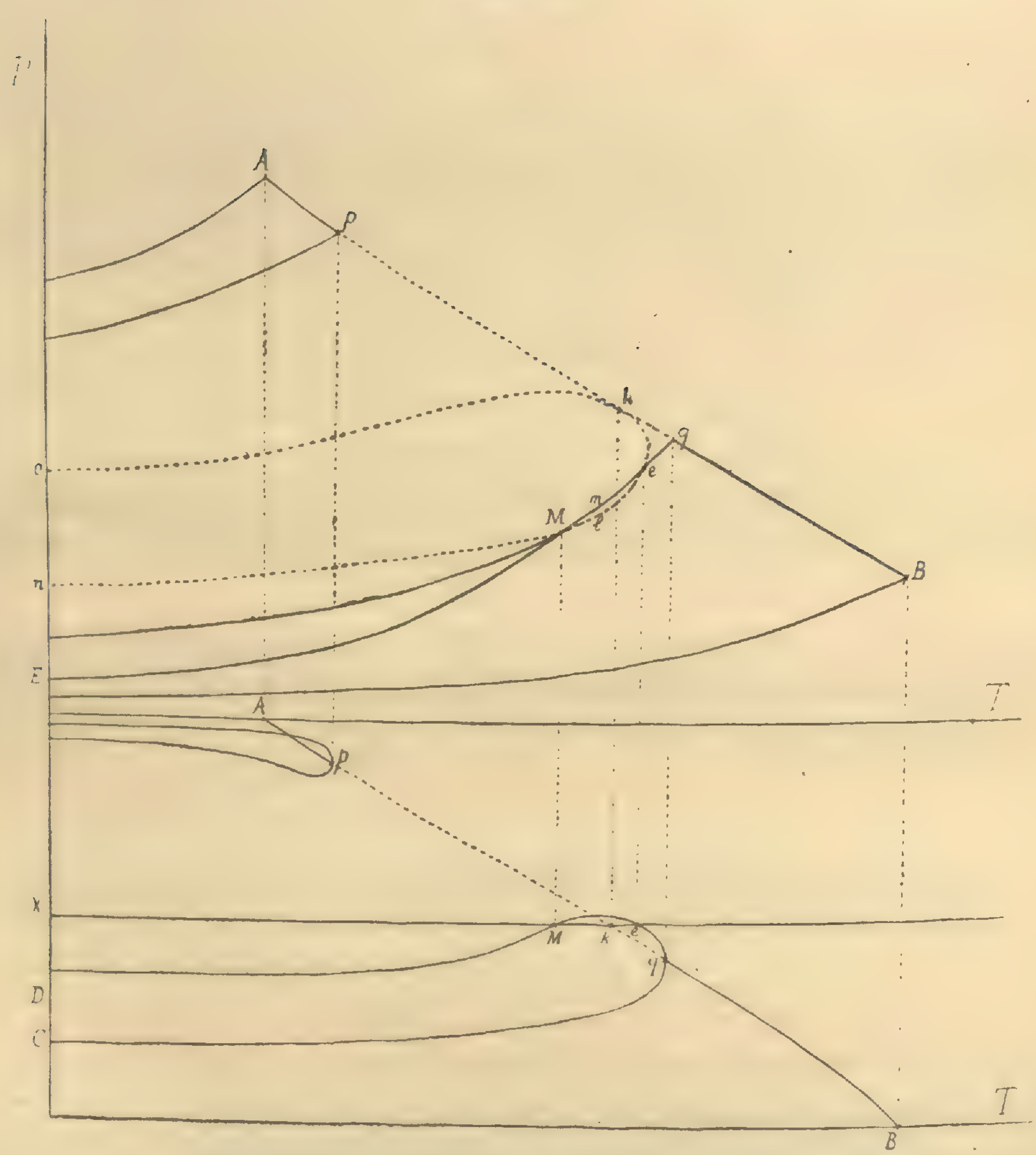

Fig. 2.

melting-point of the compound has disappeared here, that the liquid branch of the two-phase region coexisting with solid no longer cuts the line for $x = {}^1/_2$ in the T-x-projection; the vapour branch, however, meets the latter line in two points: the maximum-sublimation point M and a point e. If in this case we examine the behaviour of the compound itself, it appears that on the line EM (P-T-projection) the solid compound exists by the side of its own vapour, that on the line Mme the three phases may occur side by side, and that the region $Mmel$ indicates the area where liquid and vapour stably coexist, while a line for solid-vapour touches the three-phase line in e, and runs towards higher temperature in the P-T-projection below eq. If the solid phase does not occur, the coexistence of L and G is moreover possible in the region $nMmeko$, to which the stable region $Mlem$ then immediately adjoins.

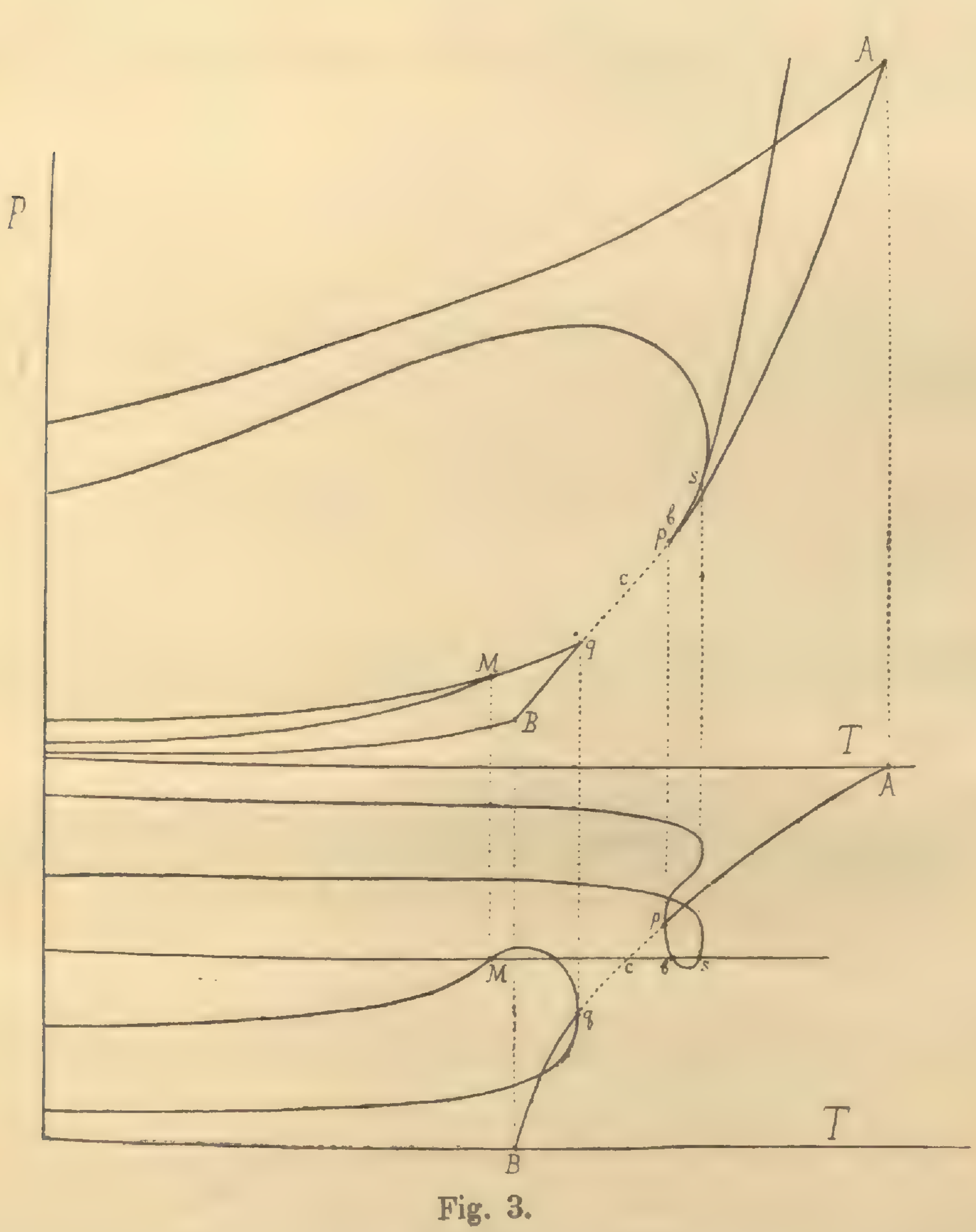

Fig. 3.

III. The two critical end-points lie on the branch SM (points p_2 and q_2 from fig. 1), so between melting point and maximum sublimation point. The liquid branch of the two-phase region of p coexisting with solid intersects the line for $x = {}^1/_2$ twice, once in the melting-point S (Fig. 3), and the second time in a point b, which lies between S and p. In the same way the vapour branch of the region of q intersects the middle phase twice, in the maximum sublimation point M and in a point lying between M and q. In b and S the compound exists by the side of liquid of equal concentration under the three-phase pressure; in the points of intersection of the vapour branch with $x = {}^1/_2$ by the side of vapour of equal concentration, with which then a liquid of other concentration coexists. The line which in this case indicates the equilibrium between liquid and gas for the concentration of the compound passes through S and M, but also through the two other points of intersection of liquid and vapour branch with $x = {}^1/_2$, and through a point c of the critical line, which is, therefore metastable there.

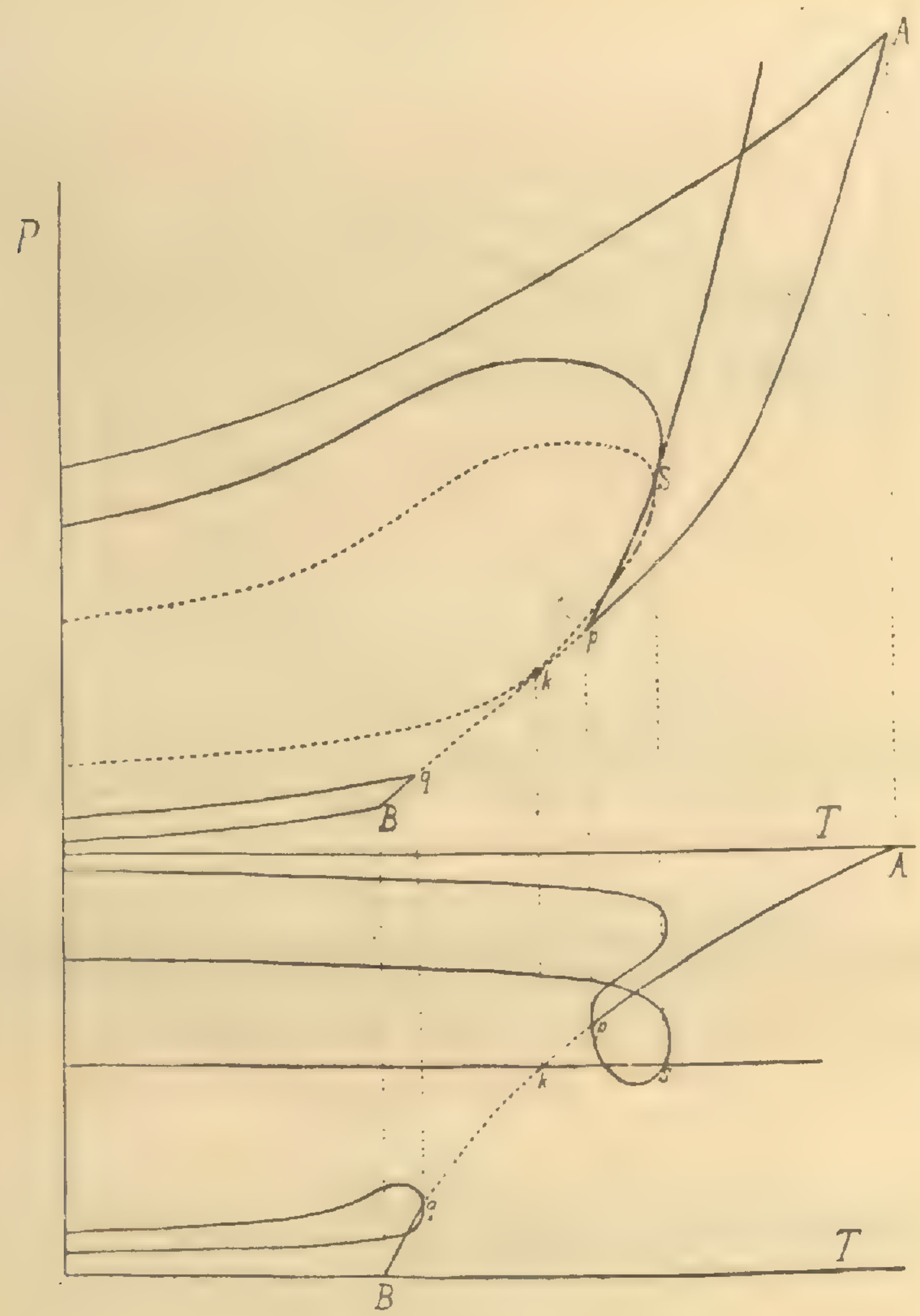

Fig. 4.

IV. One critical end-point lies on SM, the second on BM. (points p_2 and q_3 of fig. 1). In this case, which is given by fig. 4, the melting-point S still occurs on the three-phase line on the side of the first component, while the maximum sublimation point has disappeared. In connection with this is the fact that the plane of the compound is only intersected by a liquid branch; the plaitpoint k lies between p and q. In S the melting-point line touches, in the other point, where the solid compound exists by the side of liquid of equal concentration, a second line for solid-fluid, which proceeds towards lower temperature touching the three-phase line in the P-T-projection. This latter line has been omitted for the sake of clearness.

V. Both the critical end-points lie on BM (points p_3 and q_3, fig. 1) so for concentrations which lie between those of the compound and of the second component. The equilibrium liquid-gas in the middle plane, which gave rise to a metastable critical phenomenon in the preceding case, yields here a stable critical point (fig. 5). The lines that indicate the behaviour of the mixture of the concentration of the compound, have been inserted in the figure.

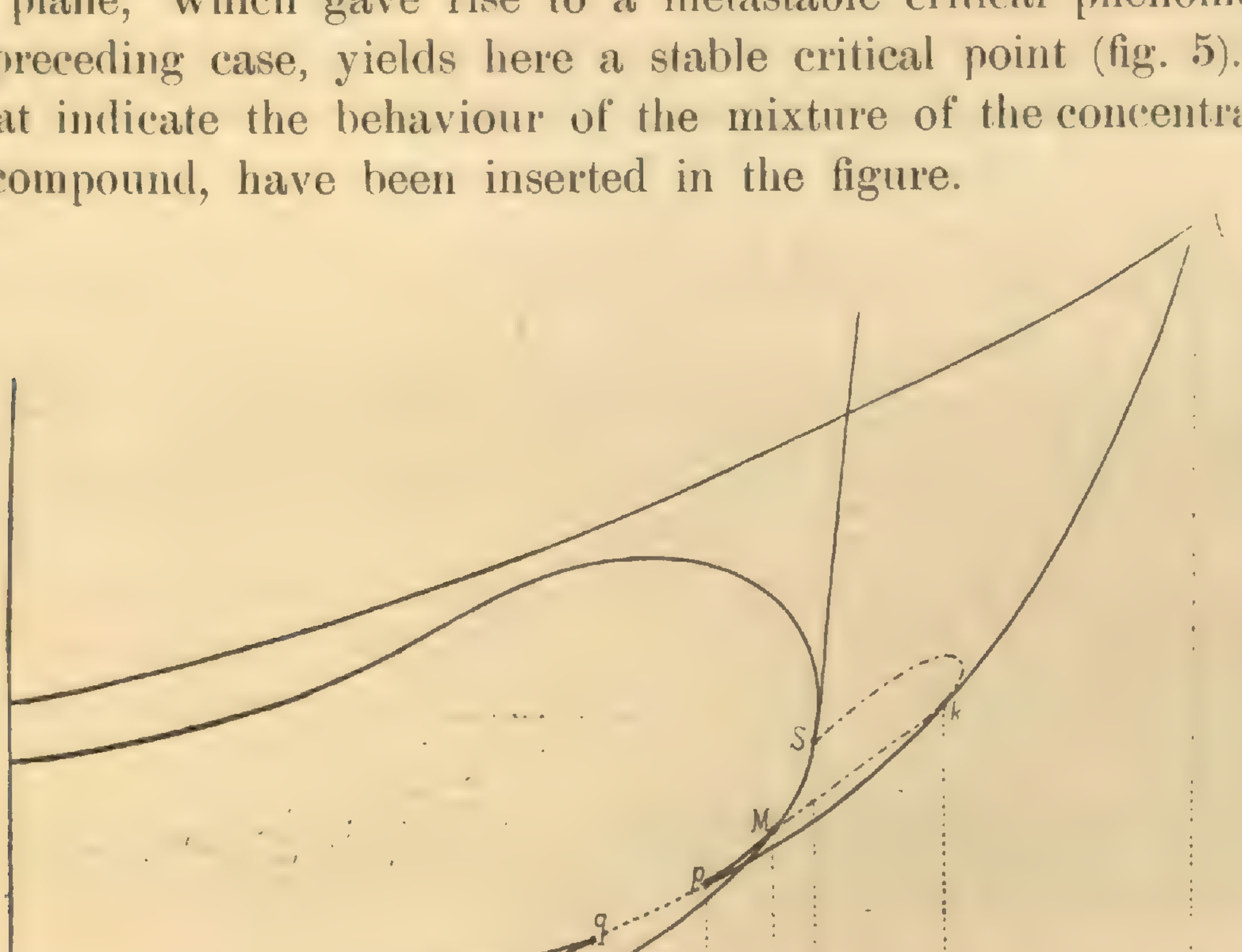

Fig. 5.

VI. One critical end-point lies on AS, the second on BM (points p_1 and q_3 of fig. 1). This case, in which both the melting-point and the maximum sublimation point have disappeared, was already discussed by Prof. SMITS in the cited paper.

The above cases, which have been derived on the assumption that the vapour pressure of the compound lies between that of the components at the same temperature, in which, therefore, the liquid-vapour surface above the melting-point gradually descends from the first to the second component, may be easily derived for systems with compounds which are less resp. more volatile than the two components, which then gives rise to the appearance of a line of minimum, resp. maximum pressure on the liquid-vapour surface. By combination of the preceding cases double intersections may occur, one of which was discussed in the cited communication.

When we consider when the cases mentioned can occur, it appears that the chance for the occurrence of case III will be the smallest, as the generally slight differences in pressure and temperature of minimum melting-point and maximum sublimation point render an intersection of three-phase line and critical line between these points improbable. So the cases II, III, and IV, where such a situation is met with, will only be rarely found, if they really occur at all; I have not succeeded in finding an instance of these cases.

The three remaining cases have this in common that they may all occur for compounds which are little soluble in one or both components. In all the systems where I have found critical end-points, this was really the case. Of the investigated systems the following belong to

Type I: Hydrochloric acid-Ammonia; Hydrogen sulphide-Ammonia; Carbonic acid-Ammonia.

Type V: Ammonia-Sulphur dioxide.

The system hydrochloric acid-ammonia, for which it did not seem improbable that type VI would hold on account of its melting-point not being known, appeared to belong to type I, as I could ascertain that when chloride of ammonium was heated a three-phase line was found which extended from about 520°—560°.

Of these systems only that of hydrogen sulphide and ammonia is suitable for a quantitative investigation of the spacial figure, though this system, too, is open to great experimental difficulties in consequence of the action of mercury on hydrogen sulphide, which is only to be counteracted with the greatest difficulty. Yet I have succeeded

in determining one critical end-point *(p)*; the second *(q)* was not to be reached on account of the pressure lying too high. Fig. 6 represents the *P*-*T*-projection of the *P*-*T*-*x*-figure. The three-phase line on

Fig. 6.

the side of the most volatile component, in our case hydrogen sulphide, practically coincides with the liquid-vapour line of the pure component; it has been drawn separate for the sake of clearness. The solubility is so slight here that the isotherm lowering of vapour-pressure (till the saturated solution under three-phase pressure is reached) is practically zero; this involves that the critical end-point *p* differs only little in pressure and temperature from the critical point of hydrogen sulphide ($T_{k_{H_2S}}$ 99.6°; T_{k_p} 103.0°; $p_{k_{H_2S}}$ 88.3 atm.; p_{k_p} 93.9 atm.).

The three-phase line on the ammonia side runs at low temperatures far below the liquid-vapour line of ammonia.

It ascends very rapidly in the neighbourhood of 110°, intersects the ammonia line in the P-T-projection, and runs almost vertical at 117°. I have been able to follow this line towards higher pressure to 225 atmospheres without any indication of a critical phenomenon. So the point q lies at a pressure which is higher than 225 atmospheres.

In the point N, which lies at about 50°, contact takes place of the minimum pressure line and the three-phase line. Hence the P-x-sections have a minimum pressure in the L-G-lines at temperatures above 50°. Of the sublimation line of the compound the situation in the neighbourhood of the maximum sublimation point was accurately determined; this latter point lies at 88.4° and 19.3 atmospheres; the melting-point could not be observed so accurately as a slight action of the mercury on the mixture in consequence of the generated hydrogen can give rise to a great error in the pressure; it lies at 116 à 117°, and at a pressure which is higher than 150 atmospheres.

While in the few cases in which the two points mentioned were accurately determined, the differences in pressure and temperature were slight, a difference occurs here of 28° and more than 130 atmospheres; this system is the first and only case in which such a situation was met with.

By the aid of closed tubes on one side, and Cailletet tubes on the other I have succeeded in determining the T-x-projection of the part of the critical line on the hydrogen sulphide side (fig. 7), and of the three-phase line on both sides (figs. 8 and 9).

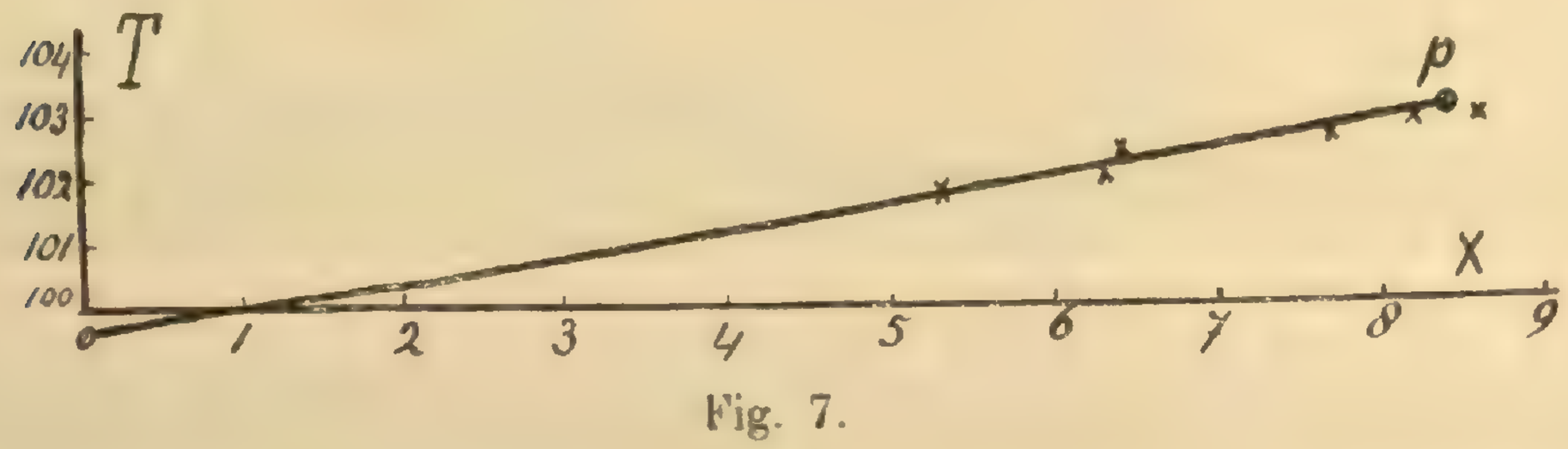

Fig. 7.

The critical line runs pretty much straight from the critical point of pure hydrogen sulphide (99.6) to the critical end-point p (103.0°, 8.4% ammonia). This line has been reproduced in fig. 8 with the liquid and vapour branch of the two-phase region coexisting with the solid compound, the former of which could be accurately determined, the latter is indicated schematically. The T-x-projection of the same region on the ammonia side (fig. 9) presents the melting-

point S (116 à 117°) at 50 % of ammonia and the maximum sublimation point M (88.4°). The liquid curve was partly determined by the method of closed tubes, partly by the Cailletet method, which yielded concordant results; the dotted part, which could not be accurately observed partly on account of the mentioned disturbing reaction, partly

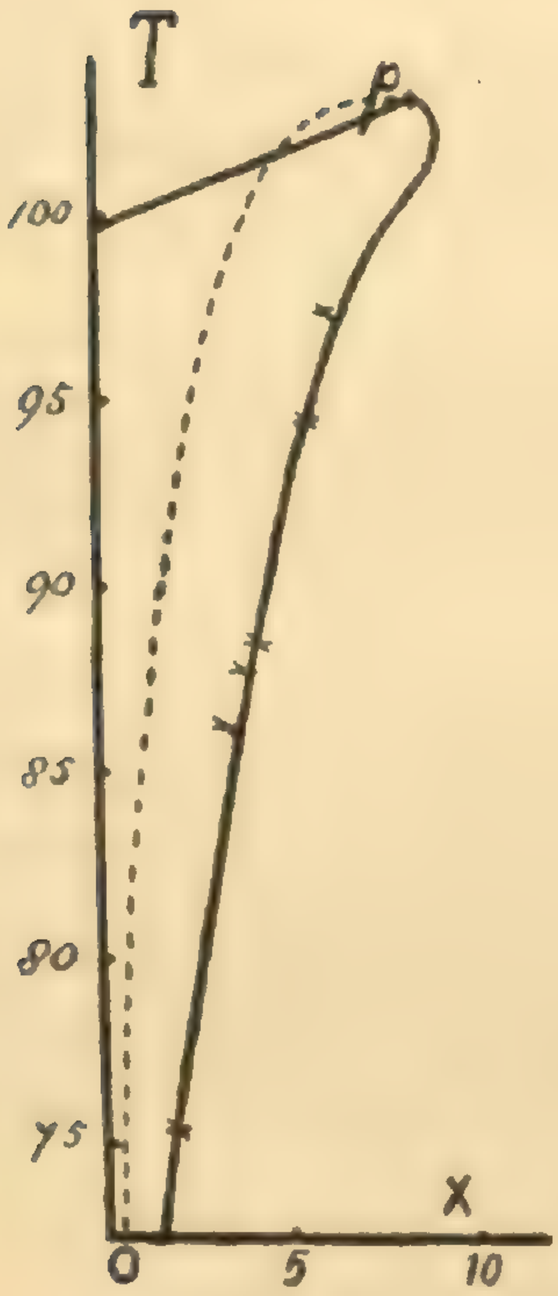

Fig. 8.

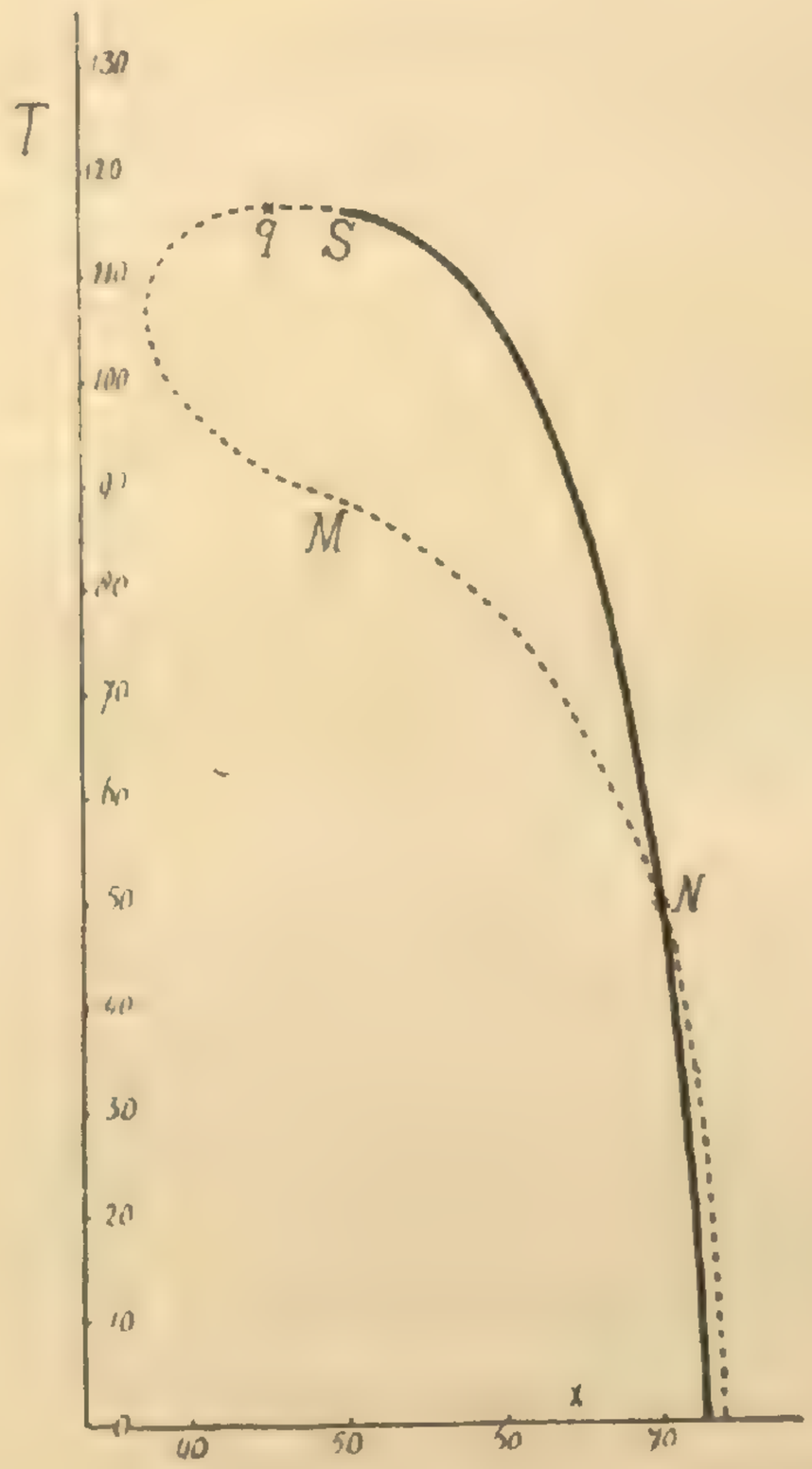

Fig. 9.

because the quantities of substance in the Cailletet tubes would have to be too small, agrees only roughly with the actual state of things with the exception of the maximum sublimation point. On the left of N the melting-point lies at higher temperature than the sublimation point, and on the other side of N the reversed situation occurs. The point N, where the two branches of the two-phase region intersect, is also to be found in the P-T-projection of fig. 6, where the minimum line of the liquid-vapour surface touches the three-phase line. With the aid of the theoretical description which Prof. SMITS gave of this case (Type I), the other lines will be clear without further explanation.

Physics. — "*Regular consequences of irregular refraction in the sun.*" By Prof. W. H. JULIUS.

The images produced on our retina, or in the focal plane of a telescope, may be considered as geometrical projections of light-emitting objects. Our idea of the object, as derived from the shape of the image, is founded on the validity of that rule. If it fails to hold, if, for instance, the rays are curved in traversing a whirling mass of air, so that they no longer form homocentric beams, the distribution of the light in the focal plane will give us a false idea of the object. What we then observe is no "image".

We cannot possibly doubt that rays, in passing through extensive gaseous parts of celestial bodies, deviate from the straight line in an irregular way. If caught in a telescope, such rays do not form an image of the source of light. Yet our conclusions regarding distribution of matter, and other local conditions in the sun, have to be deduced from the phenomena visible in the focal plane. In interpreting those phenomena we therefore are compelled to account for the probable course of the rays inside the celestial body considered.

Most investigators of solar phenomena have so far never thought this necessary. Sun-spots, faculae, flocculi, prominences, are spoken of as if they were luminous "objects" from which our telescopes show us the geometrically projected "images". This includes the *supposition* that the curving of the rays in the sun is too insignificant to have any considerable influence on the phenomena.

Astrophysicists, of course, cannot be ready to abandon their unlimited belief, that they are dealing with optical images of real objects, unless very strong evidence of the contrary be given. Indeed, at first sight such loss of confidence in familiar forms would seem to deprive us of almost every hold on the solar phenomena, and to doom all our notions to vagueness. But the step must be taken nevertheless, and we will try to show that the new way leads *not* to vagueness, but, on the contrary, to concise notions, by which the correlation of the principal solar phenomena can be clearly represented.

On the degree of refraction probably existing in the solar atmosphere.

We imagine the sun to be an incandescent mass, surrounded by a gaseous envelope, which we shall call the "solar atmosphere". Whether the latter passes gradually or abruptly into the main body, does not matter for the present. The constituents of the atmosphere

may be thoroughly mixed, but the average composition and density of the mixture will vary along the radius. *In this paper we shall only deal with the influence which the selectively absorbing mixture of gases, constituting the solar atmosphere, exerts on the white light, emitted by the lower layers of the sun.* Perhaps there exist local accumulations of separate gases: we leave that possibility out of consideration now; nor shall we notice the fact, that selective radiation by the solar atmosphere may in a perceptible degree contribute to the sun-light observed.

Besides in the direction of the radius the density is sure to vary also in other directions, and that in an irregular way, on account of the movement of the gases. The point in question is: can we reasonably admit the existence of density gradients sufficiently great to produce any considerable refraction effects?

If irregular density gradients comparable in magnitude with the vertical gradient of our terrestrial atmosphere near the surface $\left(\frac{d\Delta}{ds}=16\times10^{-10}\right)$, occur in the sun, the distribution of the light over the solar disk must already be quite perceptibly influenced thereby. Indeed, in gases we have the approximate relation [1])

$$\frac{d\Delta}{ds}=\frac{1}{R\varrho}$$

between the gradient $\frac{d\Delta}{ds}$, the refraction constant or specific refracting power $R=\frac{n-1}{\Delta}$, and the radius of curvature ϱ of rays perpendicular to the gradient. Let us suppose the average value of R in the mixture not to differ too much from 0,5, then our equation gives $\varrho=1.25\times10^{9}$ c.m.; which means, that the radius of curvature of rays, propagated at right angles with the supposed gradient, is less than one fiftieth part of the radius of the photosphere ($r=7\times10^{10}$ c.M.). Even within a relatively thin chromospheric layer the light coming from lower levels may therefore alter its direction very sensibly.

This inference holds *a fortiori* for layers, in which the pressure is several atmospheres, because there the gradients too are likely to exceed the one assumed.

And the importance of ray-curving increases greatly again with waves belonging to the nearest vicinity of absorption lines. Such waves suffer anomalous dispersion. With them the refraction constant of the absorbing gas may be several hundred times greater than

[1]) Proceedings Roy. Acad. Amst. IX, p. 352; Astroph. Journ. **25**, p. 107 (1907).

0,5, and therefore, even if that gas occupies only a small percentage in the mixture, so that its density gradients too are but a small percentage of those of the atmosphere, even then the light in question will be more strongly curved than normally refracted rays. Its radii of curvature will be very small indeed as compared with the radius of the photosphere; and much lesser gradients than the one assumed would already suffice to produce very marked refraction effects with such waves.

On the influence of a local rarefaction or condensation in the solar atmosphere on the distribution of the transmitted light.

In fig. 1 (Pl. I) SS' represents a part of a central section of the photospheric surface. Whether in that surface we meet with a physical discontinuity in the condition of matter, or not, is of little consequence in the following argument. Perfectly opaque it is not likely to be; so we may safely presume that an external point M receives more light along the direction ab than along other directions cd, ef, gh.[1]) Let an observer be stationed at a great distance in N. We imagine the atmosphere $SQQ'S'$ to have a uniform density throughout, except in the neighbourhood of M. At that point the density may be a *minimum* (case A) or a *maximum* (case B), and from there gradually pass to the uniform value on all sides. Let us suppose the gradient to be greatest in the space between the two little circles. Fig. 1 refers to case A. Rays leaving the surroundings of M in the direction MN have suffered an incurvation there, the concave side of which faces the regions of greater optical density, and, therefore, is turned away from M. From a distant point N one will observe a certain distribution of brightness, determined 1. by direction and magnitude of the density gradients, and 2. by direction and intensity of the light entering our region of unequal density. A rigorous solution would require a knowledge of the density in each point, and of the intensity corresponding to each direction of incidence; but it is not difficult to discern, that the main result must always be as follows: In the middle the rarefied region shows the intensity of the back-ground as seen in the direction fe; towards the edge m_1 the brightness will first increase, because we meet with rays emerging radially from the photosphere (direction ab); furtheron it will perhaps decrease (direction cd); but it certainly

[1]) This presumption is supported by considerations to be stated afterwards (cf. the foot note on p. 280).

must be very small near te edge m_2, from whence only rays, not coming from the photosphere, can reach the observer.

Our rarefied region, though perfectly transparent, thus looks like an object emitting at the left side less, at the right more light than the back-ground.

If in those parts of the atmosphere, which surround our rarefaction, the density were not perfectly uniform, the distribution of the brightness within the region considered would be modified, to be sure, but not so much as to change the main character of our result.

From case A we deduce as a general rule:

A relatively small region in the solar atmosphere, where the density passes through a minimum, and which, as seen from the earth, is projected excentrically on the solar disk, will show *darker* than the surroundings on the side *opposite* the centre of the disk, and may be *brighter* than the surroundings on the side *facing* the centre, the latter peculiarity however requiring, that the distance from the centre be not too small.

If the rarefaction is projected near the centre of the disk, it will show a dark rim on all sides.

Let us now consider the course of the light through a small region, enclosing a *maximum* of density (case B, fig. 2). Again the rays bend in such a way that their concave side is turned toward the places of greater optical density, that is, now, towards M.[1]) Examined from N, the region round M gives the impression as if there were an object, emitting at the left side more, at the right side less light than the back-ground.

From case B we deduce the rule:

A relatively small region in the solar atmosphere, where the density passes through a maximum, and which is seen excentrical on the solar disk, will show *darker* than the surroundings on the side *facing* the centre of the disk, and may be *brighter* than the surroundings on the side *opposite* the centre, provided the distance from the centre be not too small.

If the condensation is projected near the centre of the disk, it will show a dark rim on all sides.

In our scheme we supposed the region of smaller or of greater optical density to be approximately spherical. In order to make sure whether the above rules always represent the main character of the

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[1]) If in both cases A and B the density gradients had the same absolute values, the rays would generaliy change their direction more in B than in A, because in B they are curved in the same sense as the levels of equal density, and therefore travel a longer way through the non-homogeneous region.

distribution of brightness, also when the arrangement of the density gradients is less simple or even very irregular, we may supplement the inquiry by making a few simple experiments.

Some experiments with curved rays.

A saturated solution of common salt has at 15° the specific gravity 1.204; just as great is the specific gravity of a mixture of 78 parts of pure glycerine with 22 parts of water. A drop of one of these liquids keeps floating in the other for a considerable time, but gradually dissolves. Owing to the great viscosity of the rather strong solution of glycerine, no fast convection currents occur in this process: the chlorure of sodium and the glycerine diffuse into each other.

The index of refraction of the glycerine solution for yellow light being 1.44, that of the salt solution 1.38, the optical density will rapidly fall off in the layer where diffusion takes place. Rays may bend there with a radius of curvature of only a few millimeters. Suppose e. g. the layer in which diffusion takes place to be at a certain moment 0.02 cm. thick, then we get for the average value of $\frac{dn}{ds}$

$$\frac{1.44 - 1.38}{0.02} = 3,$$

and for the radius of curvature

$$\varrho = \frac{n}{\frac{dn}{ds}} = \frac{1.41}{3} = 0.47 \ cM.$$

So these liquids enable us to imitate the irregular ray-curving in the solar atmosphere, and thereby to preserve approximately the same proportion between the radii of curvature of the rays on the one hand, and the dimensions of the source of light on the other hand, if we only give the latter a diameter of some centimeters.

The experiments were made as follows.

Against the back of a vessel of plate-glass was placed a diaphragm with a circular opening, covered with ground-glass. A set of large strongly converging lenses formed the image of an arc light of 3 amp. at a distance of about 6 cm. behind the centre of the opening in the diaphragm. The ground-glass thus became a circular source of light of which the brightness gradually decreased from the centre towards the edge.

Let the vessel be first filled with our solution of glycerine. We force a drop of salt-solution into it from a little glass tube of which one end had been drawn out into a long and very thin capillary

(so that by plunging it into the glycerine, the latter might be stirred as little as possible), while the other end was closed with a rubber cap. The photograph α on plate II shows three drops, successively forced out of the same little tube. They appear to change their shape rather slowly. The one at the right hand is the oldest; there diffusion has proceeded farther than with the other. In the drops, solutions of different specific gravity seem to form, part of the contents moving downward, and another part upward. We observe this more clearly still in β, where three drops at a later stage are shown. Inside all these drops the optical density is a minimum, also in the tiny filaments where diffusion has proceeded far. Everywhere the rule relating to case A is fully confirmed.

Some instances of case B are represented in γ, δ, ε. The vessel was filled with the salt-solution, and a few drops of the optically denser solution of glycerine were forced into it. Owing to the smaller viscosity of the medium, the drops change and move more quickly in this case, than in the former one; and because the photographing had to be delayed until a suitable stage of the diffusion process had been reached, these drops appear very irregularly lengthened out, as a rule. But the general character of the optical phenomenon is perfectly evident in this case too: a region of greater optical density appears dark toward the centre, and bright toward the edge of the source of light. All the thin filaments even, separating from the big drops in ε, show the same peculiarity.

And we see, that near the middle of the disk a condensation as well as a rarefaction can only *reduce* the brightness. If, therefore, we introduced into our vessel a great number of drops of greater or smaller optical density, so that the medium were honeycombed with density gradients, the central parts of the luminous field would show a much lesser average brightness, than if the medium were homogeneous. Incidentally, circumstances might co-operate to reproduce by refraction nearly the maximum brightness of the source, at certain places; those places would then highly surpass the average back-ground in brilliancy.

Places considerably brighter than the average photosphere are really observed now and then, even on the central parts of the disk. This fact has been cited by FÉNYI [1]) as an argument against the idea, that the light of prominences might be refracted photospheric light, because no optical system, no reflexion nor refraction, can ever make the intrinsic brightness of an object increase. Our above expla-

[1]) Memorie della Soc. d. Spettroscopisti italiani XXXVII p. 182 (1908).

nation, however, does not clash with this law; so the objection moved by FÉNYI is refuted.

With our liquids we can easily realize both cases *A* and *B* together and at the same time. We have but to fill the vessel with a mixture of equal parts of both solutions, and then to introduce one drop of each of the original solutions: we thus obtain a region with a minimum of optical density. The photograph ζ shows on the left a drop of glycerine, on the right a drop of salt solution in their opposite optical character.

On several grounds the existence of *vortices* in the solar atmosphere is considered probable. In the axis of a vortex the density is a minimum. The refraction in the region surrounding it, must be comparable with that in our extended drops. It is supposed that sun-spots are the seat of vortices with their axes more or less perpendicular to the photosphere [1]). Now let a spot be situated near the middle of the disk. Our line of sight will make only a small angle with the axis of the whirl; the light travels a long way through the non-homogeneous region, and the effect of ray-curving must be greater than in the case of a spherical rarefaction.

In order to realize similar conditions by experiment, we only need to send the light vertically through our liquids, because the drops extend chiefly in that direction.

The same diaphragm with ground-glass was fixed in a horizontal plane, and a vessel with bottom of plate-glass placed upon it. Two mirrors, one beneath and one above the vessel, and making angles of 45° with the vertical, enabled us to arrange the illuminating and the photographic apparatus in about the same manner as before.

Some results are to be found on Plate III. The photographs η and θ were obtained with salt drops in glycerine. Even on the middle of the luminous disk may now appear very dark spots whenever the line of sight makes but small angles with the long, almost cylindrical levels of equal density. Indeed, rays emerging in the direction of the line of sight from points near the axis of that rarefaction, suffered all of them such an intense incurvation, that they must have come from the dark surroundings of the source of light.

Just as in the former set of experiments the rarefaction, if situated far from the centre of the disk, shows always a bright rim toward that centre.

With glycerine drops in salt solution it was not so easy to obtain plain-shaped vertical filaments, on account of the greater mobility of

[1]) HALE, Contributions from the Mount Wilson Solar Observatory Nos. 26 and 30; Astroph. Journ. XXVIII, Sept. and Nov. 1908.

the liquid. Yet the photographs ι and $\varkappa$ suffice to show that, with these drops too, dark spots can be produced, which, in contrast with the former case, have a bright rim on the side opposite the centre.

Some observations were made with the vessel divided into two partitions by a glass plate. In λ the left part contained the optically thinner medium with a local condensation, the right part the denser medium with a rarefaction. Finally the image μ shows both cases in *one and the same* medium: on the left two salt drops, on the right two glycerine drops are floating in a mixture of the two solutions. All these drops were lengthened in a direction nearly perpendicular to the plane of the image.

What is a sun-spot?

So far our results justify the suggestion, that the darkness of sun-spots and the distribution of brightness round about them might be mere consequences of refraction.

A difficulty arises. The typical sun-spot has a principal characteristic, which we do not observe as such in our imitations, namely: a *penumbra*, enclosing the *umbra*, from which, as a rule, it can be rather sharply distinguished. But this objection is easily removed.

Indeed, there is an essential difference between the arrangement of the gradients of optical density in a diffusing drop on the one hand, and round a vortex on the other hand. With the diffusing drop, the greatest gradients are to be found in the layer of quickest diffusion, that is, not far from the outside; whereas in the region of circulation, always surrounding a whirling mass of gas, *the gradients increase as we approach the vortex.*

It is not so easy, of course, to realize by experiment this latter case with its optical consequences; but that it must result in the appearance of an umbra and a penumbra, is clearly shown by the scheme fig. 3, plate I. Within the cylindrical space *abcd* the matter is supposed to be circulating about the vortex *W*. Rays directed towards the observer after emerging from the innermost parts of that region (the thin vortex-thread itself may be left out of regard) have suffered the greatest change of direction; they come from the space outside the photosphere, have little intensity, and produce the umbra. Parts of the spot, lying a little farther from the vortex, send us the light which left the photosphere tangentially: here we have the boundary between umbra and penumbra (in a certain sense: an inverted "image" of the solar horizon). Next follow the penumbra rays; first those having left the photosphere at great angles with the sun's

radius, and therefore being the weaker ones; then, as we approach the outer limits of the region *abcd*, the rays that come gradually straighter from the source.

A disturbance of this "normal" arrangement of density gradients round a vortex may incidentally cause the photospheric light to follow a shorter path towards the observer, even through the central parts of the spot. The brilliant tongues and bridges, often observed in spots, find a natural explanation in this way.

Within a streaming mass of gas the velocity usually changes periodically (think of gusts of wind!); so there are shells of greater and of smaller density following each other alternately, and cutting the stream-lines at right angles. In the circulating matter round a vortex such shells must assume almost radial directions, but at the same time be slightly curved in a spiral shape. Refraction in an optical system thus constituted, brings about a distribution of brightness, known to us as the radial-filamentary structure of the penumbra [1]).

We do not want, in our spot theory, to make any special supposition as to the form of the region in which the density is a minimum. If it resembles a cylinder with its axis pointing towards us, the refraction effects will indeed be somewhat greater than if it were, for instance; spherical; but in the latter case too they may be strong enough and will bear the same principal character. Our explanation of the umbra and the penumbra rests on the only supposition, that in a certain region of the sun's atmosphere the density gradients are sufficiently great [2]), and gradually decrease from a central point outwards.

Anomalous dispersion in the region of a sun-spot.

So far we only dealt with the effects of refraction in general, without noticing the disparity of its values with the various waves of the spectrum, that is, the *dispersion*.

In general the refraction-constant of transparent media is positive; in the visible spectrum it increases with decreasing wave-length. Substances, however, showing absorption lines, may have refraction constants of such extreme variety of magnitude with the waves

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[1]) The spiral-shaped flocculi visible on HALE's spectroheliograms with H_α (Astroph. Journ. XXVIII, Sept. 1908), and covering much greater regions, probably have a similar origin.

[2]) Astronomers who do not even hesitate to admit violent eruptions in sun-spots, and irregular velocities of many kilometres per second as a rule all over the sun's surface, will certainly not think the density gradients, required in our argument, unreasonably great.

belonging to the nearest vicinity of the absorption lines, that the above regular dispersion vanishes if compared with this anomalous dispersion. Waves lying on the *red*-facing side of absorption lines, and very near to them, we shall denote by R-light in this paper; neighbouring waves lying on their *violet*-facing side, we shall call V-light. With the R-light the refraction constant has rapidly increasing *positive* values as we approach the line; with the V-light it has rapidly increasing *negative* values as we approach the line.

The refracting power $n - 1$ of a gas is equal to the product $R\Delta$ of its refraction-constant into its density.

ARAGO and BIOT found that the refracting power of a mixture of gases equals the sum of the refracting powers of the constituents:

$$R_m\Delta_m = R_1\Delta_1 + R_2\Delta_2 + etc.$$

As with most kinds of light all gases have a positive R, the refracting power $R_m\Delta_m$ of the mixture is in general positive. But with V-light $R_m\Delta_m$ may be negative, because that term of the sum, which bears upon the constituent producing the absorption line in question, may with V-light have a negative value so great, as to surpass the sum of the remaining, positive, terms. It is presupposed, of course, that the Δ of the said constituent is not too small, or in other words: the vibrating system, producing the line which we consider, should not be too scantily represented in the mixture.

Near lines of *principal* constituents the anomalous refraction predominates to such a degree, that the negative refracting power of the mixture with V-light is, in absolute magnitude, relatively little inferior to its positive refracting power with R-light.

What are we to conclude from these facts with regard to the spot-spectrum?

Let us first consider the case, that a region with a minimum of density is found just in front of the middle of the disk. Waves with which $R_m\,\Delta_m$ is positive, especially those belonging to the much refracted R-light, will in that region deviate conformably to the scheme of our drops of salt solution; the V-light, on the other hand, with which $R_m\,\Delta_m$ has great negative values, will in the same system of gradients deviate in the opposite way, that is, corresponding to the scheme of the glycerine drops. Both the R-light and the V-light are more weakened by dispersion than the rest of the spectrum: the Fraunhofer lines must in general be widened or winged in the spot-spectrum.

As the wings are caused by refraction, not by absorption, the

spot-lines are not properly "widened absorption lines", but rather absorption lines, embedded in *dispersion bands* [1]).

Not all Fraunhofer lines are widened or strengthened in the spot-spectrum. The individual differences of behaviour I hope to refer to at a later date. For the present we have only to remember the fact, that the composition of the mixture of gases cannot be the same at all levels of the solar atmosphere. It therefore also depends on the level in which the local rarefaction occurs, which lines of the spot-spectrum will be enveloped in strong dispersion bands.

We shall now treat of the case that a rarefaction is projected on the solar disk at a certain distance from the centre.

The R-light must show a distribution of brightness reminding of that which we observed in our experiments with salt drops, that is: it is strengthened at the side of the spot, turned towards the centre of the disk, and very weak at the opposite side.

The V-light is curved the other way; the distribution will resemble that shown in our experiments with glycerine drops, that is: it is strengthened at the side, turned away from the centre, and very weak at the side, facing the centre of the disk.

These rules do not involve any special supposition as to the arrangement of the density gradients with respect to their magnitude. We may therefore make our idea of the manner in which the R-light and the V-light are distributed in an excentrically placed spot, more precise, by again introducing our previous assumption that in the spot-region the gradient decreases from the interior ontwards. As a simplification we suppose that region to be nearly spherical.

Our figures 4 and 5 show the paths of the R-light and the V-light respectively. (Waves, not suffering anomalous refraction, will conform to the scheme fig. 4, as with them $R_m \Delta_m$ is positive, but the degree of refraction will be small; so they will produce a spot of which the umbra is smaller than the one represented in fig. 4). We see that with R-light the umbra is shifted towards the limb, whereas at the opposite side of the spot, in c, a bright place appears, because there emerges the light which left the photosphere perpendicularly. With the V-light, on the contrary, the umbra is shifted towards the centre, and the brightest light, emitted along the normal of the photosphere, emerges from p.

Two spectroheliograms of the same typical spot, simultaneously

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[1]) Proc. Roy. Amst. VII, p, 134—140; IX, p. 343—359.
Astrophysical Journal **21**, p. 271—291 (1905) and **25**, p. 95—115 (1907).

made with the R-light and the V-light of a suitable line [1]), should show the above difference very distinctly.

The spectroscope will reveal the following particulars.
Suppose the slit to be so placed in the sun's image, that it bisects the spot and coincides with a radius of the disk. In fig. 6 PC be a Fraunhofer line; P points to the periphery, C to the centre. The horizontal bands indicate the spectrum of the umbra and the penumbra as shown with light not undergoing anomalous refraction. With the R-light the umbra is greater and extends chiefly towards P (cf. fig. 4); so in the upper half of the spectrum the Fraunhofer line appears to be shifted to the red, and more markedly still, because at the same place in the spot the V-light attains its greatest brightness (cf. the point p in fig. 5). In the lower half of the spectrum R-light and V-light have changed parts [2]).

The widening of the line extends beyond the average penumbra because with R- and V-light the penumbra too is greater than with waves not subject to anomalous dispersion.

We conclude that in the spectrum of an excentrical spot — the slit bisecting that spot in the direction of the centre of the disk — the Fraunhofer lines must be more or less curved in the shape of the letter S, to a degree, depending on the intensity of the spot's distance from the centre of the disk. Whenever that distance is so

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[1]) Perhaps some iron line will serve the purpose better than one of the winged calcium or hydrogen lines, whose refraction effects might be confusingly great.

[2]) The distribution of the light in fig. 6 has been deduced from that in figures 4 and 5, in which process the shape of the dispersion curve, as given in fig. 8, is paid due attention to.

Indeed, one must imagine the whole spot of fig. 4 to appear smaller and smaller as the waves we are considering recede from the absorption line, because in fig. 8 the ordinates $(R_m\,\Delta_m)_R$ gradually decrease towards the red, until they reach the constant value $R_0\,\Delta_0$ (cf. p. 281). The distributions of the light along a cut PC through fig. 4, such as they would successively appear with increasing wave-lengths, were plotted side by side in fig. 6, going from the absorption line to the red.

On the violet side of the line one has to proceed in the same way. There the negative ordinates of fig. 8 decrease as we recede from the line, and the distribution of the light prevailing along a cut PC through fig. 5, must be transferred to fig. 6 on a smaller and smaller scale. But in the point c' of fig. 8 we have $(R_m\,\Delta_m)_V = 0$; light of that wave-length suffers no refraction at all in the solar atmosphere, so that the spot-spectrum is here interrupted. Next, $(R_m\,\Delta_m)_V$ attains positive values, increasing up to $R_0\,\Delta_0$; the distribution of the light has again to be taken from fig. 4.

So the spot-lines are not only inclined, but also asymmetric, a feature which especially the weaker lines will show, provided that the dispersion of the spectral apparatus be sufficiently high.

small, that there is no sufficient asymmetry in the supply of light, the inclination of the lines becomes imperceptible.

With any other direction of the slit the effect must be less; t vanishes when the slit bisects the spot in a direction at right angles to a line joining the spot and the centre.

Is it necessary to admit radial movement in sun-spots?

Bulletin N°. XV of the Kodaikánal Observatory (Febr. 1909) contains a remarkable communication by J. EVERSHED, entitled: "Radial movement in sunspots". The new facts there described chime perfectly and in every detail with the above necessary consequences of refraction. In the spectra of all the spots examined Mr. EVERSHED found the lines displaced according to the following law. Wherever a spot be situated on the solar disk — provided that the distance from the centre surpasses 10' — the majority of the Fraunhofer lines of its spectrum are slightly inclined when the slit bisects the spot in the direction toward the centre of the disk. The inclination is less with other directions of the slit, and disappears when the slit is at right angles to the sun's radius. The displacement of the lines is always to the red on the side of the spot, turned towards the limb, and to the violet on the side facing the centre; it differs in magnitude with the individual lines [1]).

To explain these phenomena Mr. EVERSHED invokes DOPPLER'S principle. He attributes the displacements to a radial movement outwards from the spot-centre. The motion must be essentially horizontal, or parallel to the sun's surface; the velocity seems to increase, from the centre of the spot outwards, yet at the limits of the penumbra the motion apparently ceases abruptly. The lines of the true umbral spectrum seem usually to be almost entirely unaffected by motion in the line of sight. So there is not a kind of spring in the umbra, and we are puzzled with the question, whence the matter comes, which is permanently spreading out in the spot, and where it accumulates. Moreover, the maximum velocities indicated by the

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[1]) H_β was not inclined. We presume that the anomalous refraction produced by the hydrogen of the solar atmosphere is *too considerable* to become apparent in the same way as the refraction caused by other elements. Even in the average solar spectrum the dispersion band of H_β is more than one Ångström unit wide; in the spotspectrum it may perhaps extend so much farther, that one is inclined to look upon it as not belonging to the line, and, therefore, to consider the line as weakened. Now, the narrow central line is the real *absorption* line H_β, which of course cannot be displaced or curved by refraction.

displacements of different lines range from 0 to more than 2 kilometers per second, so that the elements would move independently. And Mr. EVERSHED himself remarks, that the hypothesis of a radial movement in spots seems entirely out of harmony with the conclusions arrived at by Prof. HALE in his researches on vortices and magnetic fields in sun-spots.

To my mind, the difficulties involved in Mr. EVERSHED's explanation of the important facts which he has discovered are insuperable. Motion in the line of sight cannot be the cause of those systematic displacements. Neither can they be accounted for by pressure or by the influence of a magnetic field, without introducing very improbable hypotheses *ad hoc*. On the other hand I could not find any serious difficulty in describing the newly-discovered phenomena as mere consequences of the anomalous refraction of photospheric light in local rarefactions of the solar atmosphere.

On the origin of the Fraunhofer lines.

We considered a sun-spot as a region in which the density gradient regularly decreases in all directions from the centre outwards, thus preserving the same sign unto the outer limit of the penumbra.

Density gradients of the same order of magnitude are supposed to occur just as well outside the spots, at many places throughout the solar atmosphere, but there they are not arranged by the systematizing influence of an important vortex, and often change their sign.

Now if the widening of the Fraunhofer lines in the spot-spectrum must be ascribed to anomalous dispersion in extensive regions containing a deep "depression", why should not the width of the lines in the ordinary solar spectrum also be caused for a considerable part by dispersion in minor rarefactions and condensations?

At first sight the evidence seems unfavourable to this view. For one might contend that, if the R-light and the V-light were not really *absorbed* in the solar atmosphere, but only dispersed, such waves must leave the sun after all, be it at points very distant from those where they emerged from the photosphere, and could not, therefore, be wanting in the *average* sun-light.

But this argument is incorrect.

To understand this, imagine the whole solar atmosphere divided by surfaces into alveoles or cells in such way, that each cell encloses either a maximum or a minimum of density. The shape of the cells will vary to the extreme.

Let us first consider a nearly spherical cell enclosing a *minimum* (fig 7). All the light which, coming from the photosphere SS', enters

this region in a direction, making an angle φ with the normal to the photosphere, will leave the cell as a diverging beam. The spreading is wider in proportion as the refraction constant R_m for the kind of light considered is greater. (If R_m is negative, the rays will first converge, but afterwards diverge just as well).

With small values of φ and moderate refraction, all the incident rays will leave the cell in such directions, that they continue to move away from the photosphere. But with larger values of φ as well as of R_m there is increasing chance, that some of the dispersed rays are directed back to the photosphere and thus prevented from leaving the sun.

The total quantity of light entering our cell is found by integrating round the normal, and with φ between the limits 0 and $\frac{\pi}{2}$. (It is true that into our cell some light will enter, which comes from higher levels, where it has been refracted downward by other cells, but that contribution is small in comparison with the supply from beneath). All this light would be emitted by the celestial body if there were no incurvation of rays; but now a part returns to the photosphere. That part is greater in proportion as R_m has greater (positive or negative) values.

The cells are not spherical indeed, but it is clear that if other shapes are considered, our result must on the average continue to hold. And a cell enclosing a *maximum* of density, makes the rays first converge, then diverge if R_m is positive, and makes them always diverge if R_m is negative.

So *all* the inequalities of the density co-operate to prevent a certain part of the light of the photosphere from passing through the solar atmosphere. [1])

In the solar spectrum the waves, which belong to the nearest vicinity of the absorption lines, appear with less intensity than the other waves, not because they are absorbed in the solar atmosphere, but because they are partly directed back to the interior. The Fraunhofer lines are absorption lines, enveloped in dispersion bands.

Asymmetry of the Fraunhofer lines.

From our modified conception of the solar spectrum — which only supplements KIRCHHOFF's interpretation — it follows that all

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[1]) The same refraction effect must also cause the brightness of the disk to decrease from the centre to the limb, even if perhaps the cosinus-law holds for the radiation of the photosphere, and independent of a general absorption which the solar atmosphere might exert. (cf. the footnote on p. 268).

Fraunhofer lines must show certain systematic distinctions as compared with the character of pure absorption lines.

Dispersion bands, produced by the solar atmosphere, must be asymmetrical, the narrower ones to a higher degree than the wider ones.

Indeed, we may represent the refracting power of that atmosphere for each kind of R-light appertaining to a given line, by the expression:

$$(R_m \Delta_m)_R = (R_1 \Delta_1)_R + (R_0 \Delta_0)_R$$

in which the term $R_1 \Delta_1$ relates to the constituent of the atmosphere producing the line in question, and $R_0 \Delta_0$ means the refracting power of the mixture of the remaining constituents. Both terms are positive.

In a similar expression, concerning the V-light of the same line:

$$(R_m \Delta_m)_V = (R_1 \Delta_1)_V + (R_0 \Delta_0)_V ,$$

the term $(R_0 \Delta_0)_V$ will not differ to any appreciable degree from $(R_0 \Delta_0)_R$; but now R_1 is negative. At equal distances on either side of the absorption line, R_1 has (according to the law of anomalous dispersion) nearly equal values of opposite sign. If, therefore, we select on the violet side of the line a wavelength, matching the first-chosen R-light symmetrically, we may write:

$$(R_m \Delta_m)_V = -(R_1 \Delta_1)_R + (R_0 \Delta_0)_R ,$$

so that, with such pairs of wave-lengths, the relation holds:

$$(R_m \Delta_m)_R + (R_m \Delta_m)_V = 2 R_0 \Delta_0 = const.$$

Suppose we approach the line from both sides in a symmetrical way, then $(R_m \Delta_m)_R$ and $(R_m \Delta_m)_V$ begin with being positive; one increases, the other decreases, and when $(R_m \Delta_m)_R$ has reached the value $2 R_0 \Delta_0$, $(R_m \Delta_m)_V$ passes zero, and is going to take increasing negative values. But in *absolute magnitude* $(R_m \Delta_m)_V$ will always be inferior to the corresponding $(R_m \Delta_m)_R$; the difference grows from 0 to $2 R_0 \Delta_0$, and then remains constant when we approach nearer to the absorption line.

Figure 8 serves to elucidate this. The ordinates a and a', b and b' etc. represent values of $R_m \Delta_m$, bearing upon waves that are symmetrically situated with respect to the absorption line. The ordinates of the dotted line have the nearly constant value $R_0 \Delta_0$.

The degree to which the light is dispersed by refraction in the solar gases, is determined by the *absolute magnitude* of $R_m \Delta_m$; so the average effect must be greater on the red side of an absorption line than on the violet side. The asymmetry of the dispersion band must manifest as a displacement of the Fraunhofer line towards the red. If however the terms $\pm R_1 \Delta_1$ are highly predominating in a rather wide band of the spectrum, in other words, if the vibrating systems which give rise to our line constitute an important part of

the solar atmosphere, the asymmetry, and with it the displacement, become imperceptible.

A displacement of the majority of the Fraunhofer lines towards the greater wave-lengths, amounting to a few thousandth parts of an Ångström unit, was discovered by JEWELL as early as 1896. But there were numerous exceptions: lines showing no displacement, or a displacement towards the violet (with respect to their positions in the emission spectrum produced in the laboratory).

Recently M. M. FABRY and BUISSON [1]) applied their fine, extremely accurate interferential method in comparing iron lines of the arc-spectrum with those of the solar spectrum. Only narrow lines could be studied in that way. Their results confirmed those obtained by JEWELL: in general displacement to the red, but many exceptions. The latter, however, appeared to bear exclusively upon lines which under ordinary circumstances are not very sharp in the arc-spectrum, and are widened asymmetrically when the current is increased. With such lines the localisation in the emission spectrum had perhaps not been so accurate as could be desired. FABRY and BUISSON succeeded [2]) in making these dubious lines perfectly sharp, by producing the iron-arc in vacuo. On comparing the solar spectrum with the arc-spectrum thus obtained, they found, that the exceptions had disappeared: all the lines examined were shifted to the red in the Fraunhofer spectrum. The displacements ranged from 0,005 to 0,010 Ångstr. units with lines whose width varied between 0,07 and 0,16 units (intensity 1 to 8 according to ROWLAND's scale).

At the beginning of their interesting communications FABRY and BUISSON remark: "Le déplacement des raies permet (donc) de mesurer les variations de pression. En comparant les raies du spectre solaire avec les raies correspondantes de l'arc à la pression atmosphérique, on a un moyen d'évaluer la pression de la couche renversante, si toutefois aucun autre phénomène n'intervient." Supposing that there were no other disturbing phenomena except those which they had excluded in their experiments, they calculate that the average pressure in the reversing layer must be from 5 to 6 atmospheres.

We hold the opinion that this valuation is certainly too high. Indeed, the observed displacements cannot have been exclusively produced by pressure, because refraction is sure to bring about line-shifts in the same direction. And if perhaps future research might induce one to think it probable, that at the same level of the sun the pressure

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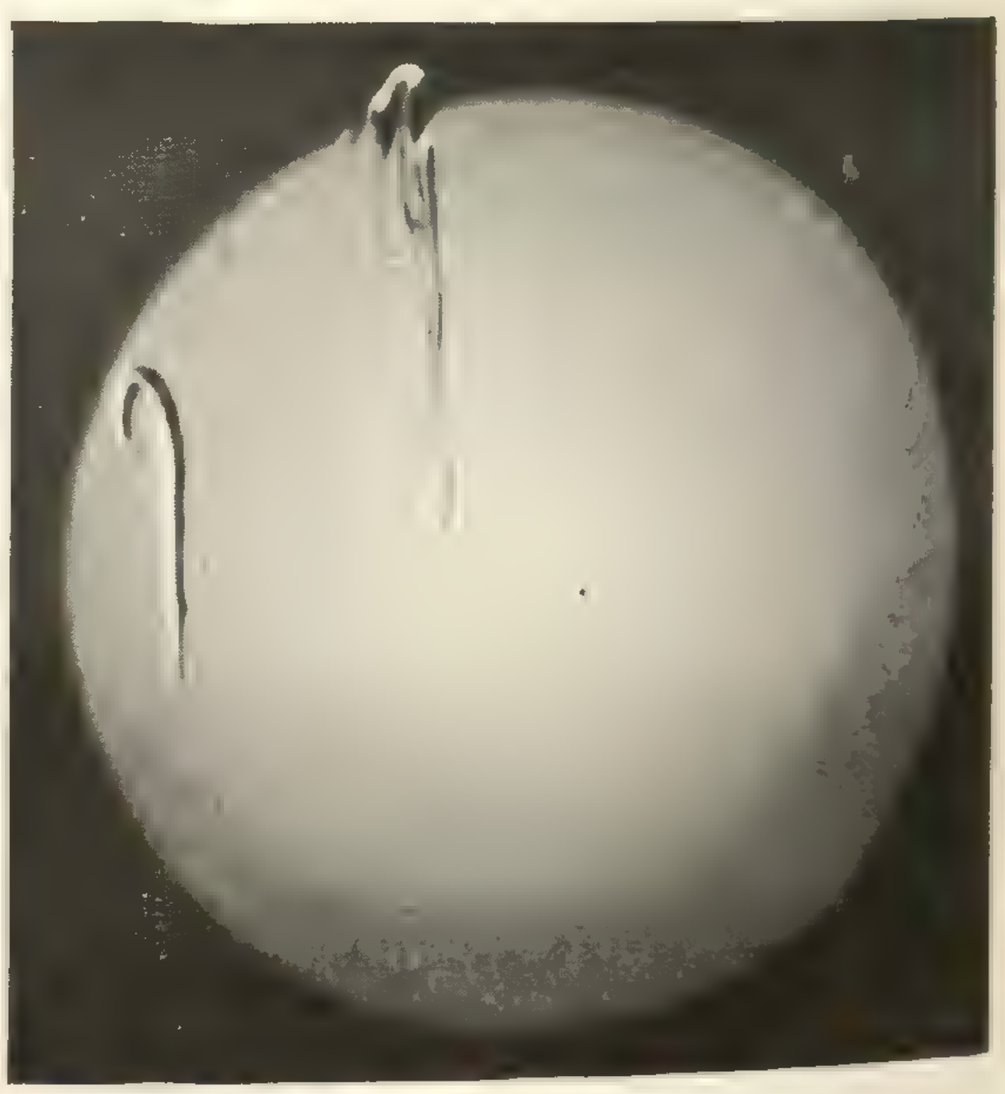
[1]) FABRY et BUISSON, Comptes rendus, 15 mars et 29 mars 1909.

[2]) FABRY et BUISSON, Comptes rendus, 10 mai 1909.

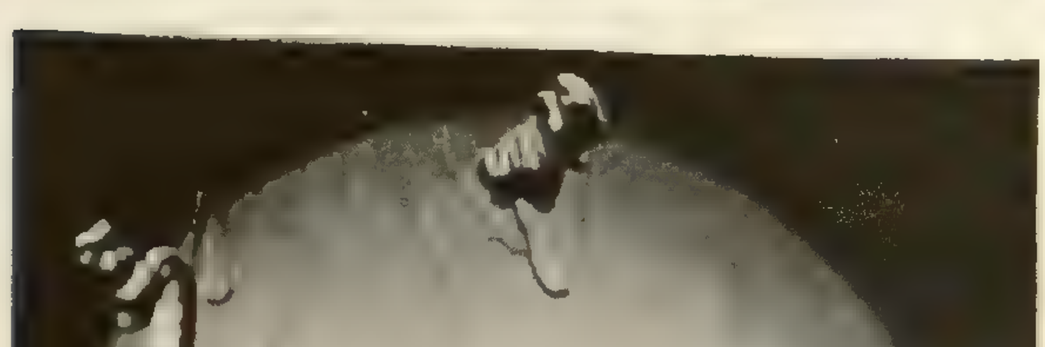
Plate II.

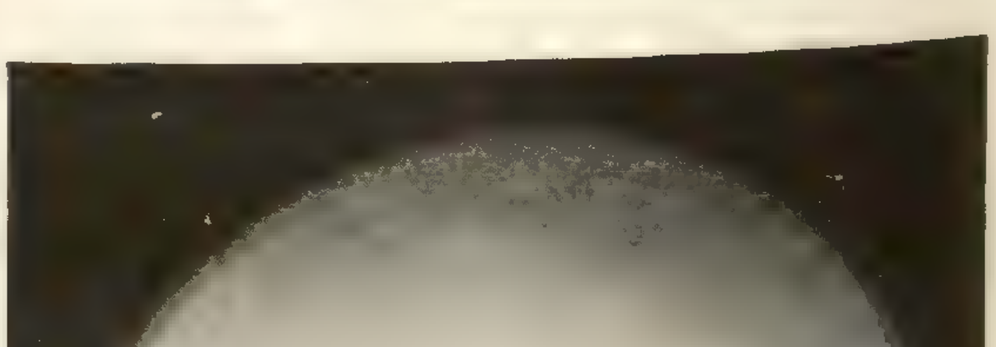
α

γ



ε

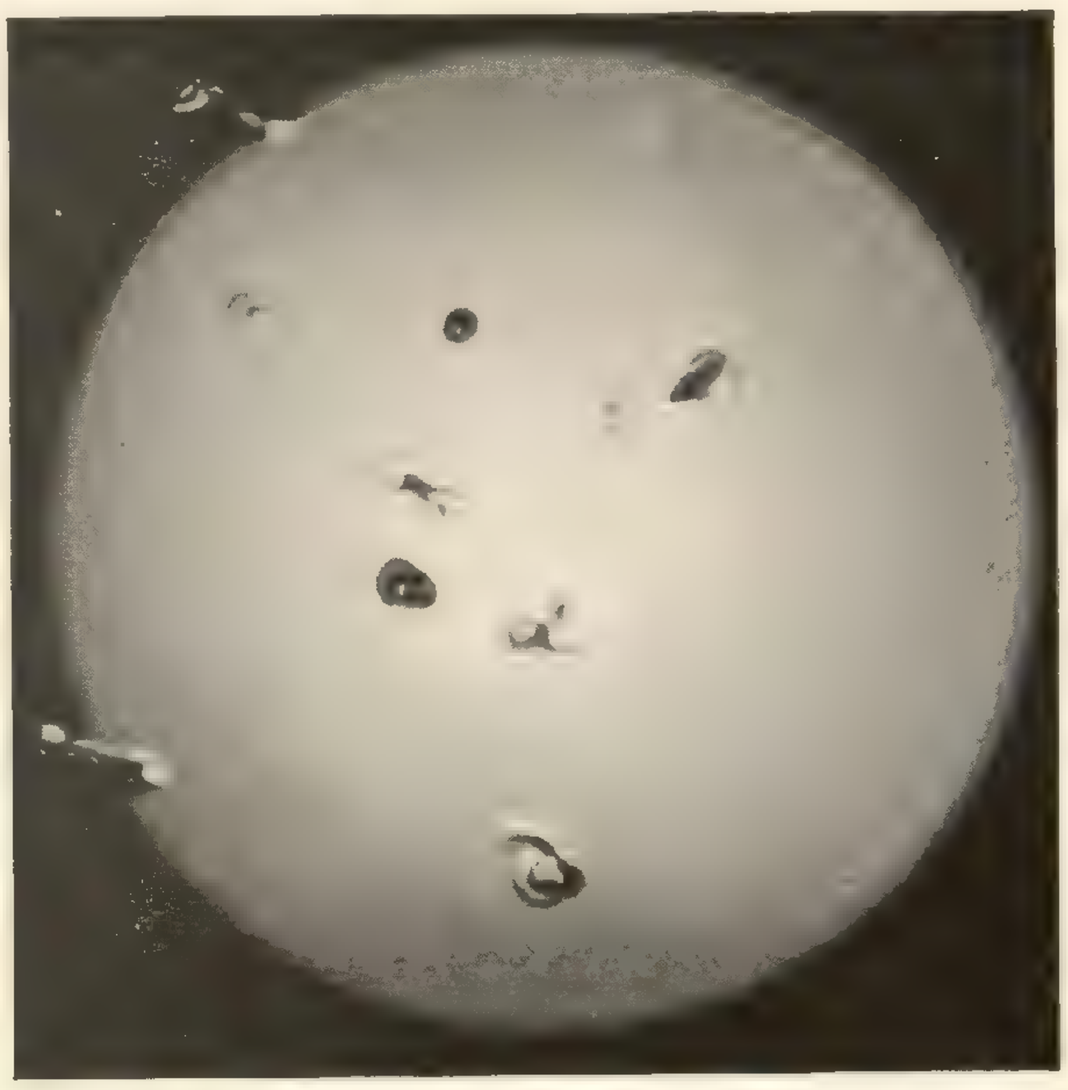




W. H. JULIUS. "Regular consequences of irregular refraction in the sun."

Plate III.

η



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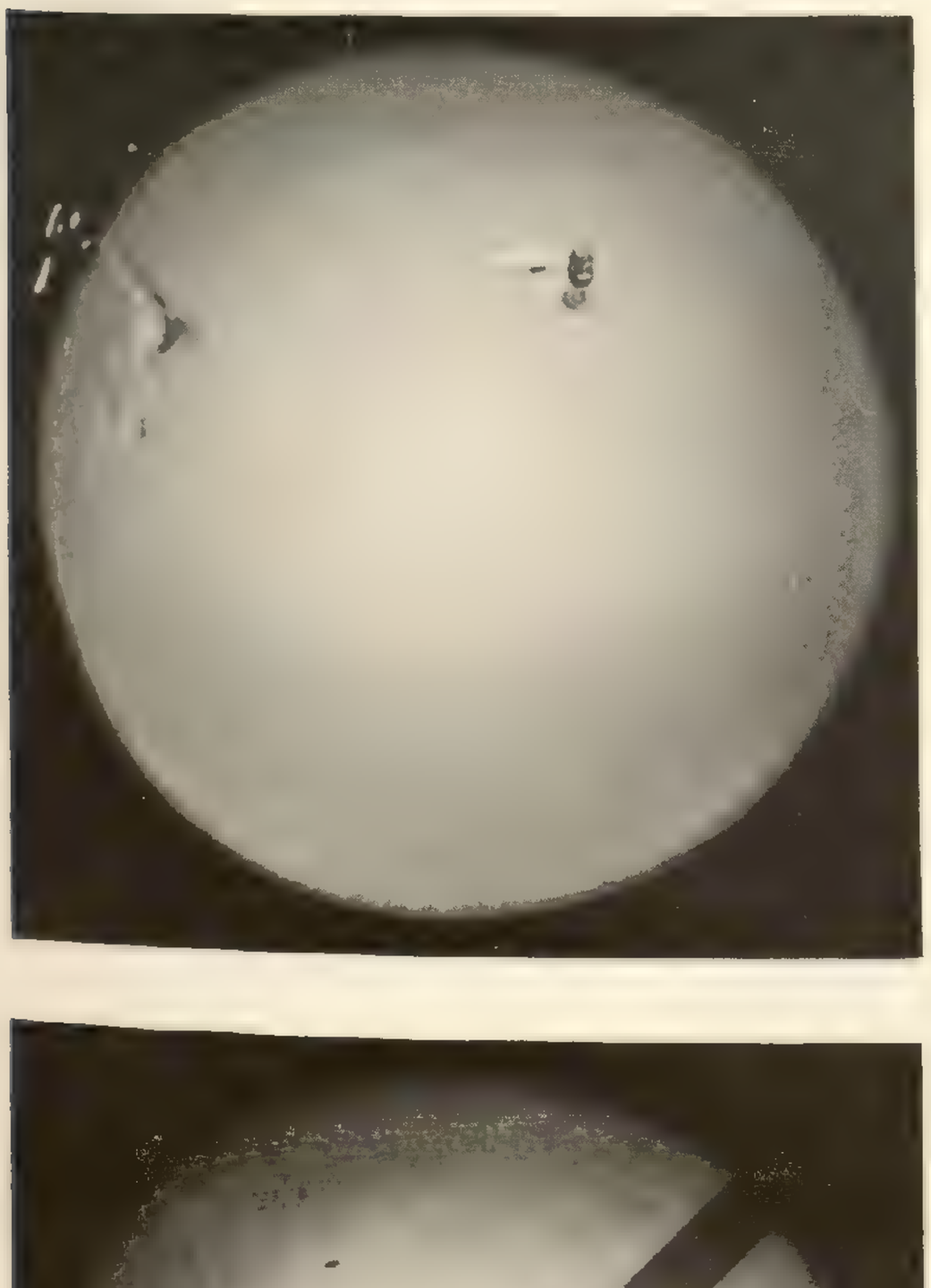
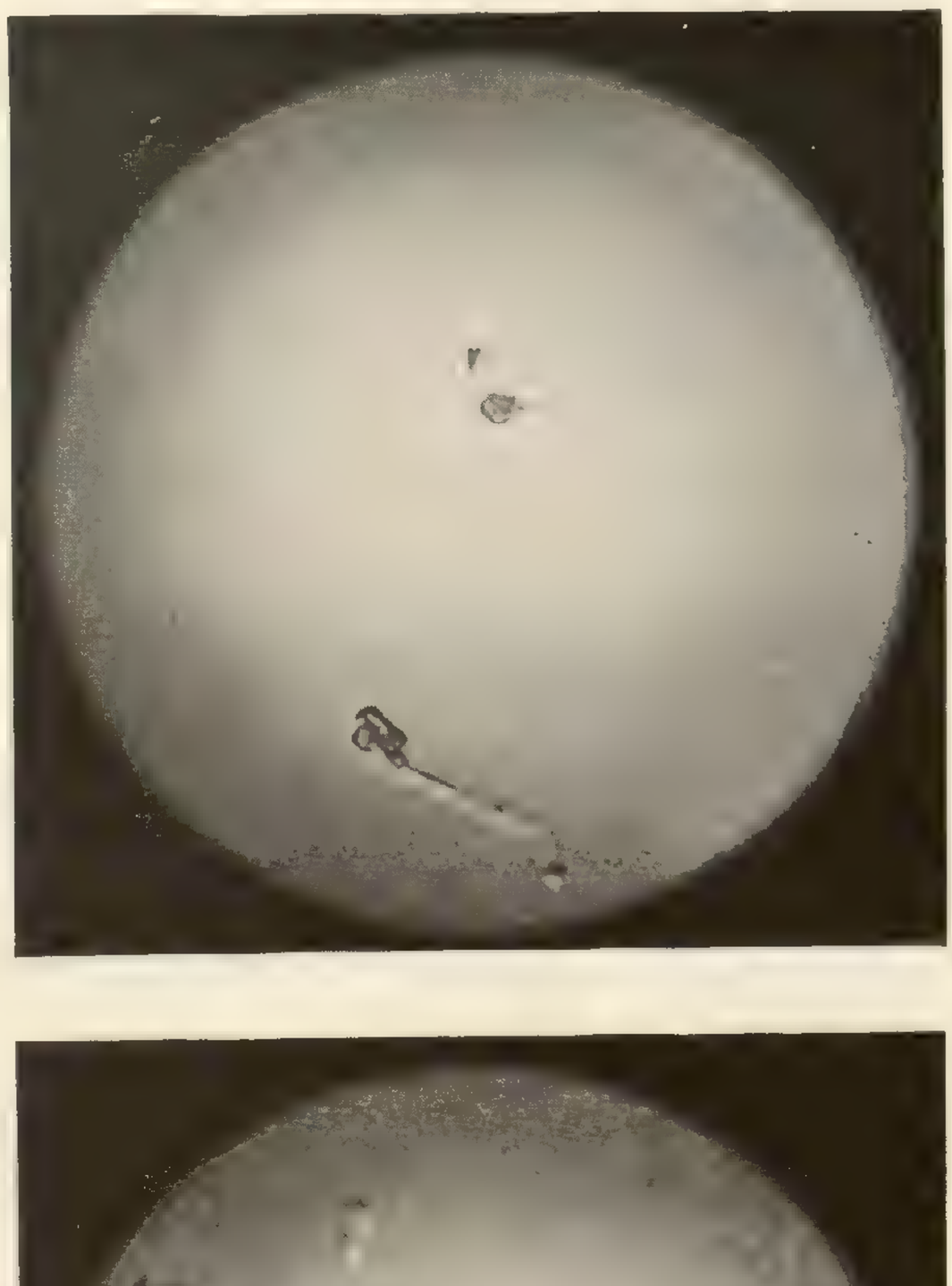
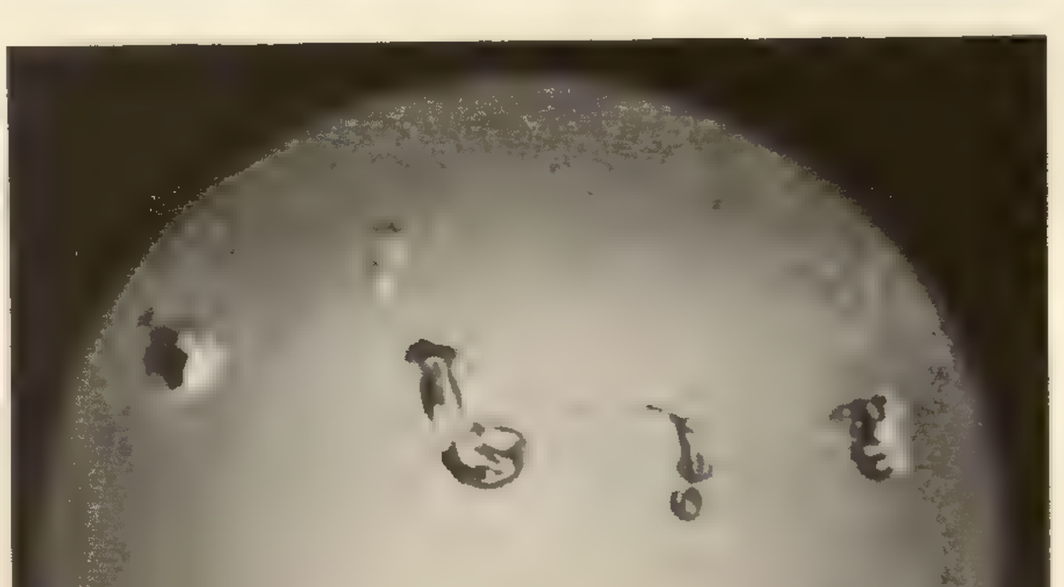


ι



κ

λ



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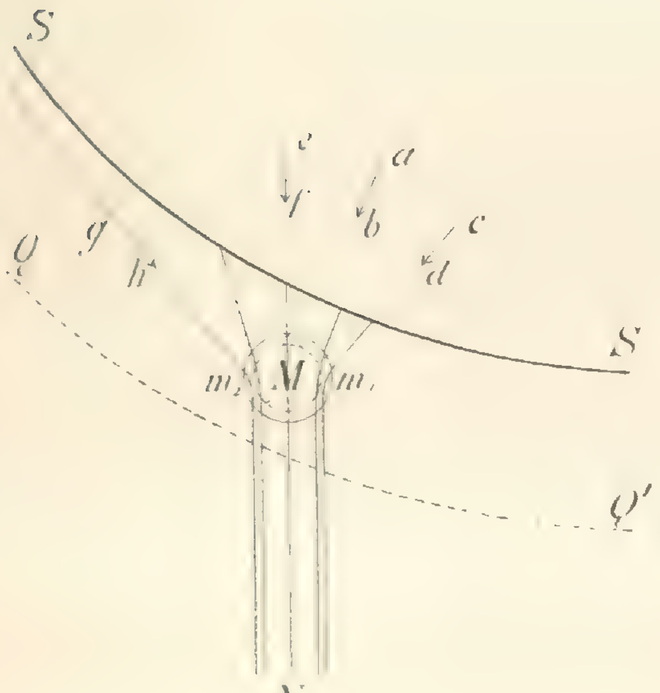

Fig 1.

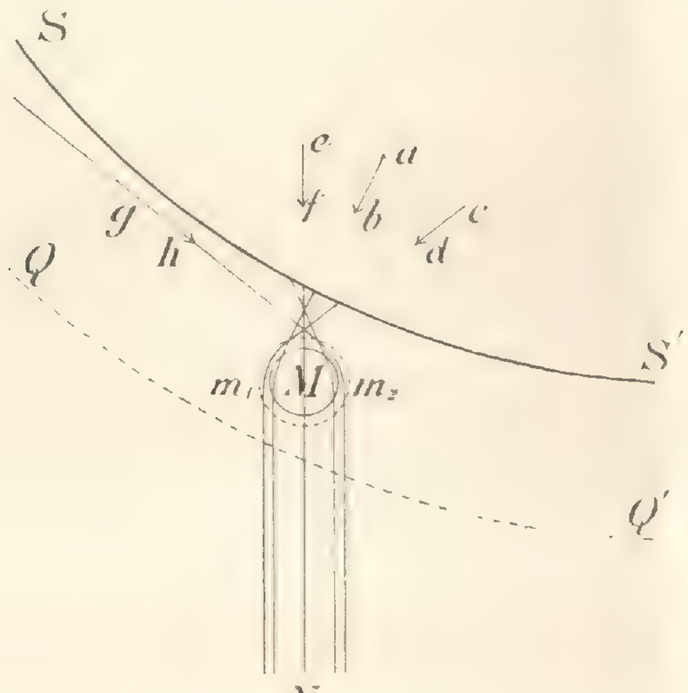

Fig. 2.

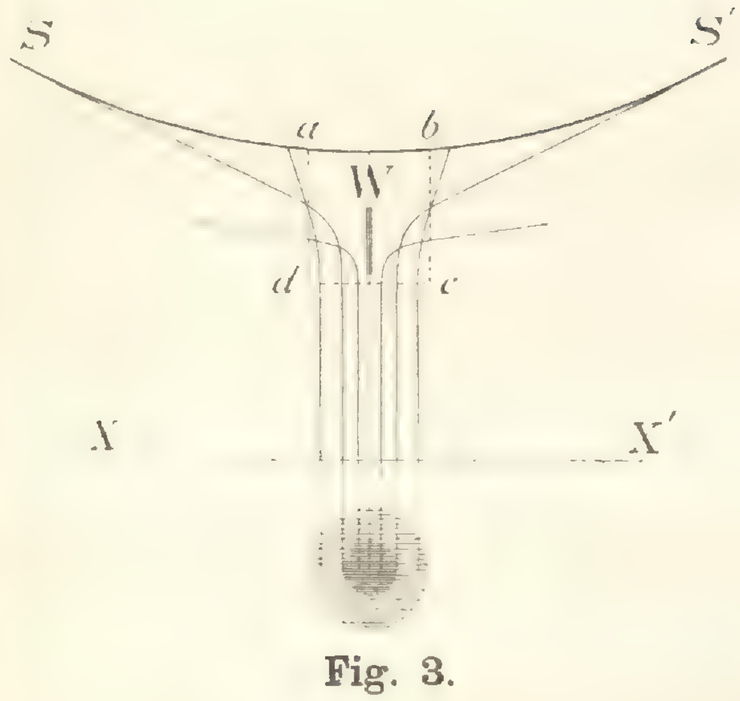

Fig. 3.

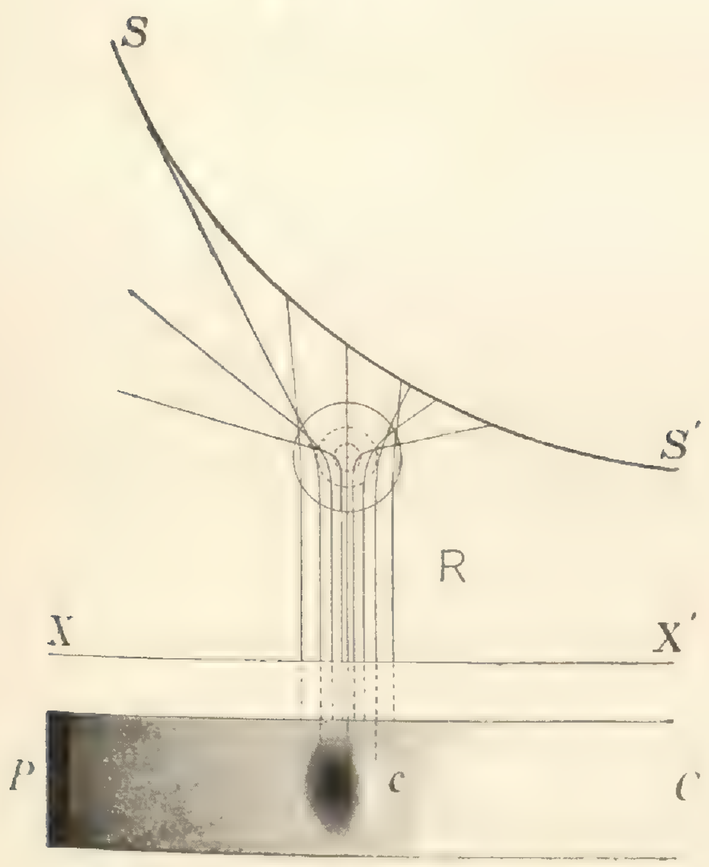

Fig. 4.

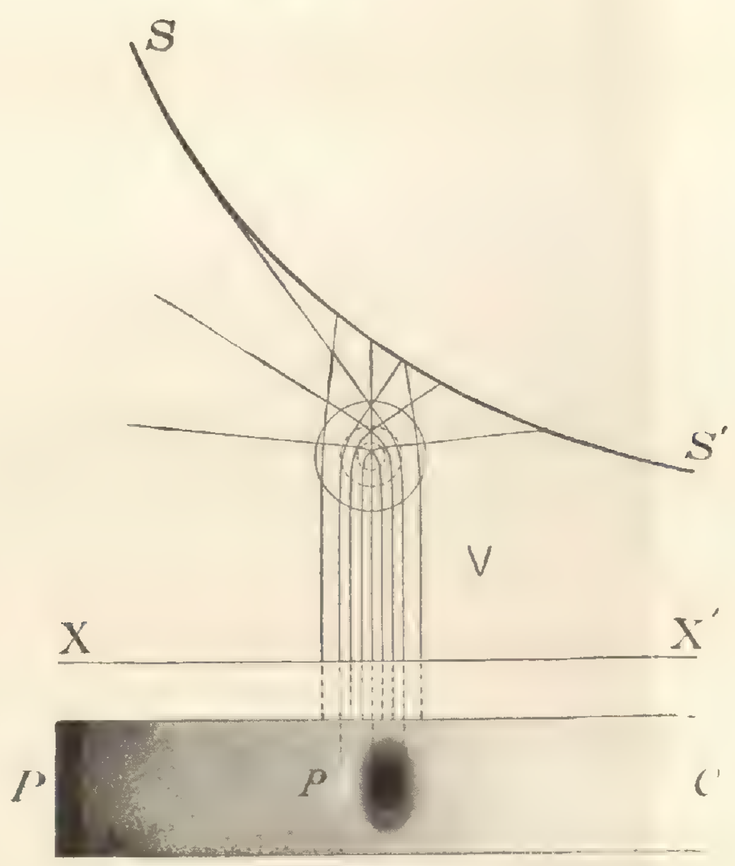

Fig. 5.

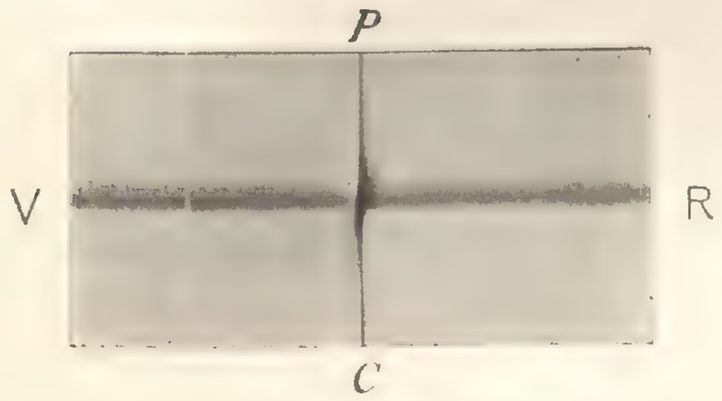

Fig. 6.

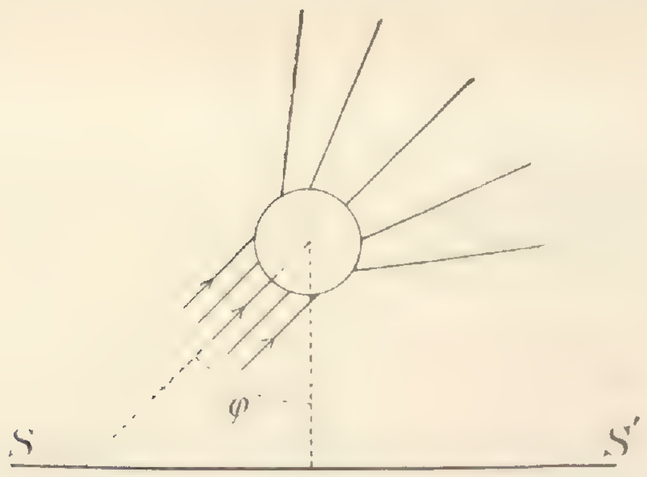

Fig. 7.

Fig. 8.

is *very* much smaller than 5 or 6 atmospheres, there will be no incompatibility with FABRY and BUISSON's observational results, because anomalous dispersion can be made responsible for the *whole* effect.

Similar considerations apply to another general result of recent observations, viz. to certain systematic differences existing between the spectrum of the centre and that of the limb of the disk.

BUISSON and FABRY resume the facts as follows[1]):

1. Il y a un changement d'aspect pour certaines raies. Pour les fortes raies, la pénombre est affaiblie dans le spectre du bord. Parmi les autres raies, quelques-unes sont renforcées ou affaiblies sur les bords du disque, et le changement est en général de même sens que dans le spectre des taches[2]).

2. HALM a annoncé[3]) que certaines raies subissent, du centre aux bords, un léger accroissement de longueur d'onde; ses mesures ont été faites sur deux raies rouges du fer, en comparant leurs positions à celles de deux raies telluriques voisines. Il a trouvé un déplacement de 0,012 angström.

Using their interferential method BUISSON and FABRY examined into 14 lines of various metals (intensities 2 to 6 on ROWLAND's scale), and found HALM's result confirmed: a displacement to the red, varying between 0,004 and 0,006 Ångström units, when passing from the centre to the limb. (Two vanadium lines were not displaced). Moreover, those same lines appeared to be about 0,010 Ångström units wider at the limb than at the centre of the disk. So we may take it as if the widening affects chiefly the red-facing side of the lines.

If one wishes to interpret this phenomenon too by means of the pressure effect — as the discoverers did — one must admit that deeper layers, in which the pressure is higher, contribute relatively more to the formation of the absorption lines near the limb than near the centre of the solar disk.

We will not dwell on various difficulties, encountered in developing this idea; probably it will be possible to remove them by introducing additional hypotheses concerning the thickness of the layers, or the temperature and other physical conditions of the absorbing vapours.

At all events it is unnecessary to ascribe the above differences between the spectra of the central and the marginal parts of the disk entirely to pressure; for anomalous refraction — which undoubtedly comes into play — acts exactly in such a way, that in the

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[1]) BUISSON et FABRY. Comptes rendus, 28 juin 1909.
[2]) HALE and ADAMS. Astroph. Journ. **25**, June 1907.
[3]) HALM, Astronomische Nachrichten, nos. 4146—47, 1907.

spectrum of the limb a widening of narrow lines, especially on their red side, is to be expected.

Indeed, rays coming from the limb have, as a rule, accomplished a longer distance through the solar gases than rays coming from the centre, and, therefore, were more subject to loss of intensity by the process of incurvation towards the photosphere. The amount of the irregular ray-curving depends on the absolute magnitude of $R_m \Delta_m$, which (near the weaker lines of the solar spectrum) is sensibly greater with R-light than with V-light. So the lines must chiefly widen at their red-facing side, in proportion as the opportunity for losing light increases.

That the character of the limb-spectrum resembles that of the spot-spectrum in many respects, is immediately explained by the refraction theory. Inded, the light which we receive from the inner zones of the penumbra, has followed paths, along which the conditions of limb-rays prevail, (cf. fig. 3).

On the other hand, there are numerous peculiarities in the behaviour of various Fraunhofer lines, which we cannot so easily explain by simply applying the laws of refraction — either because entirely different processes are involved, or perhaps because the consequences of refraction have to be more deeply scrutinized. (We remind of the weakening of the wings of the stronger lines in the limb-spectrum; of the weakening of several lines in the spot-spectrum; of the different way in which the hydrogen lines behave in the spectra of the limb and the spots, etc.). All this requires an ample study of details, in which it is necessary to dispose of an extensive collection of observational data.

Finally I wish to call attention to an inference deduced from our conception of the Fraunhofer spectrum, which, to my knowledge, is not yet supported by any direct observation.

Among the various kinds of V-light of each line, one kind must exist, for which $R_m \Delta_m = 0$ (at c' in fig. 8). Rays of this and of neighbouring wave-lengths suffer less refraction than rays, belonging to blank parts of the spectrum; so they must produce strips of greater brightness (pseudo-emission lines) in the spectrum of the centre of the disk. With strong Fraunhofer lines the point c' lies far away, on a part of the dispersion curve so slightly inclined, that the bright band must be vague and inappreciable; but with narrower lines the bright companion will also be narrow, and relatively more conspicuous. Its distance from the absorption line is always smaller than the width of the Fraunhofer line. By applying the extremely sensible method of FABRY and BUISSON the bright lines may perhaps be detected.

(October 28, 1909).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Saturday October 30, 1909.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van Zaterdag 30 October 1909, Dl. XVIII).

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Mathematics. — "*Continuous one-one transformations of surfaces in themselves.*" (2nd communication).[1]) By Dr. L. E. J. BROUWER. (Communicated by Prof. D. J. KORTEWEG.)

(Communicated in the meeting of June 26, 1909).

We shall now consider an arbitrary twosided[2]) surface and we shall submit it to an arbitrary continuous one-one transformation in itself with invariant indicatrix.

Under a *limit region* of the transformation we shall understand a region of the surface lying entirely outside its image region, but losing that property by any extension.

However it may happen that a limit region allows of *enlargement*, i.e. can be united after an indefinitely small modification of its boundary with a finite adjacent region into a new limit region, whose surface, measured by a certain system of coordinates, is then of course greater than the old one's. This will be illustrated by the following developments.

Under a *transformation domain* we shall understand a limit region not capable of enlargement, and our intention is to construct such a transformation domain.

To this end we start from two arcs of simple curve[3]), which are each other's image, which have two and not more than two points in common, and which do not cross each other in those points. We shall suppose, that these arcs have no endpoint in common; their situation with respect to each other then still allows of various possibilities, indicated by fig. 1.

With the aid of these two arcs we now construct two regions G and G', bounded by simple closed curves, which regions are each other's image and lie entirely outside each other, whilst their boundaries have two arcs of simple curve in common. In fig. 2 this has been executed for the second possibility indicated by fig. 1.[4])

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[1]) See these *Proceedings*, Vol. XI, page 788.

[2]) A onesided surface falls under our result only when brought into a continuous multivalent correspondence with a twosided one.

[3]) i.e. "einfache Kurvenbogen" after SCHÖNFLIES. In my preceding communications of this year (these *Proceedings* XI, p. 788, 850) I translated "einfach" by "single". Finding the term "simple closed curve" used by VEBLEN, I shall adopt in future this mode of expression.

[4]) Only in the fifth and eighth case of fig. 1 this might give rise to some difficulty, namely if the two common points of the arcs are each other's image. By a slight modification of the figure this difficulty can then be cancelled.

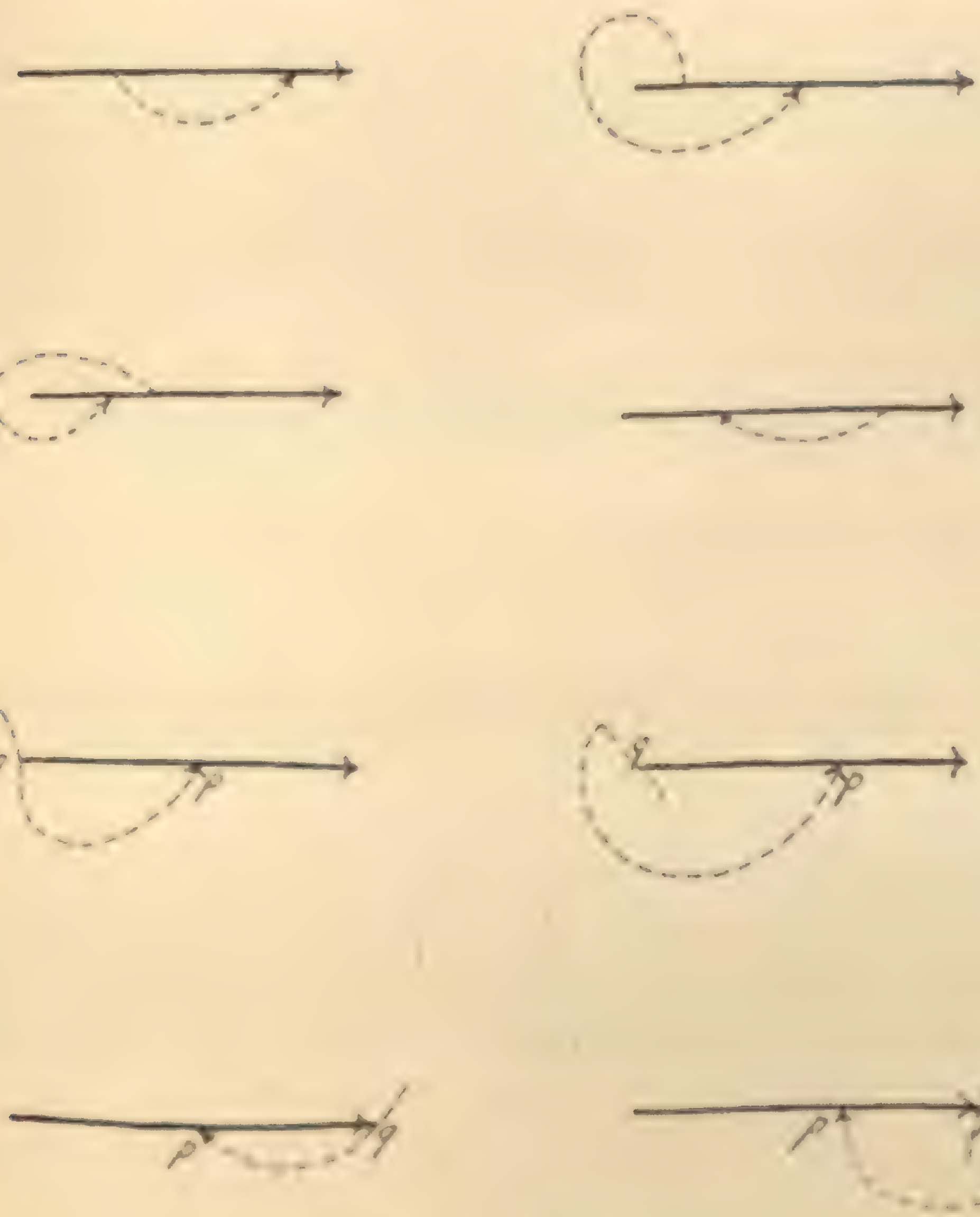

Fig. 1.

Fig. 2.

On the boundary of each of the two regions G and G' then directly lie some [1]) arcs of simple curve, which will remain *accessible*.[2])

[1]) In the first two cases of fig. 1 this number is 4; in the sixth it is 4 or 5; in the seventh 3 or 5; in the eighth it is 5.

[2]) I.e. "erreichbar" after Schönflies, Bericht über die Mengenlehre II, p. 176.

for any limit region, to which the region G may be extended, and likewise for its image region.

We now extend G in such a way, i. e. we replace it in such a way by a region in which it is contained, that, when G' has obtained the corresponding extension, the extended regions G and G' still lie outside each other. We repeat this extending process so often, until we get a limit region, and this will be the case after a denumerable number of extensions.

If possible, we then, after an indefinitely small modification of the boundary, execute an enlargement of this limit region; by this its property of being a limit region will in general be lost, but can be regained by a denumerable number of new extensions. This new limit region we again try to enlarge, and in this way go on, until by a denumerable number of operations a *transformation domain* O is obtained.

If in the surface no region exists, which at once with all points of its boundary is invariant for the transformation, the domain O can at most determine two rest regions, namely a rest region R_1, in which O' lies, and a rest region R_2, identical to R_1 or not, in whose image region R'_2 lies O.

If namely a third rest region G_3 existed, G_3 as well as G''_3 would be free of O, as well as of O'. Let P be an arbitrary point on the boundary of G_3 not coinciding with its image P'. Let us construct about P and P' simple closed curves, chosen as small as one likes, which are each other's image and bound regions π and π', then $\mathfrak{m}\ (O, G_3, \pi)$ and $\mathfrak{m}\ (O', G''_3, \pi')$ each contain one of a pair of regions, which are each other's image, and which one can make to contain of O and G_3 resp. of O' and G''_3 as closely as one likes *approximating* [1]) partial regions, to which O and O' might be *enlarged*, but this would clash with the property of a transformation domain. The same reasoning holds with a slight modification for the supposition that G_3 and G''_3 coincide.

We shall say, that a region of a surface *does not break the connection of the surface*, if it determines only one rest region, possessing for analysis situs the character of a rest region of a trema. Then of course the region itself is singly connected.

We shall now assume, that O does not break the connection of the surface. Then it is bounded by a *closed curve* [2]) K, which on account of the commencement of the construction according to fig. 2 is a *non-singular closed curve*, and therefore does allow of

[1]) Schönflies, Bericht über die Mengenlehre II, p. 104 sqq.

[2]) Id., ibid., p. 118 sqq.

division into two proper arcs, and not into two improper ones [1]).

We shall now consider on each of the curves K and K' the cyclically ordered sets u and u' of their outwardly accessible points. These determine on each other certain segments σ_n and σ'_n, forming in pairs certain cyclically ordered sets $\varkappa_n$ of points accessible from a rest region of O and O', as is schematically illustrated by fig. 3.

If we make a circuit along u and u' together in such senses, that segments σ_n and σ'_n are reached simultaneously, this circuit is made *in opposite senses* for u and u'.

So if we make a circuit along u and u' together in such a way that points corresponding for the transformation are reached simultaneously, then with respect to the former order they must meet twice in that course, and at such a meeting either corresponding

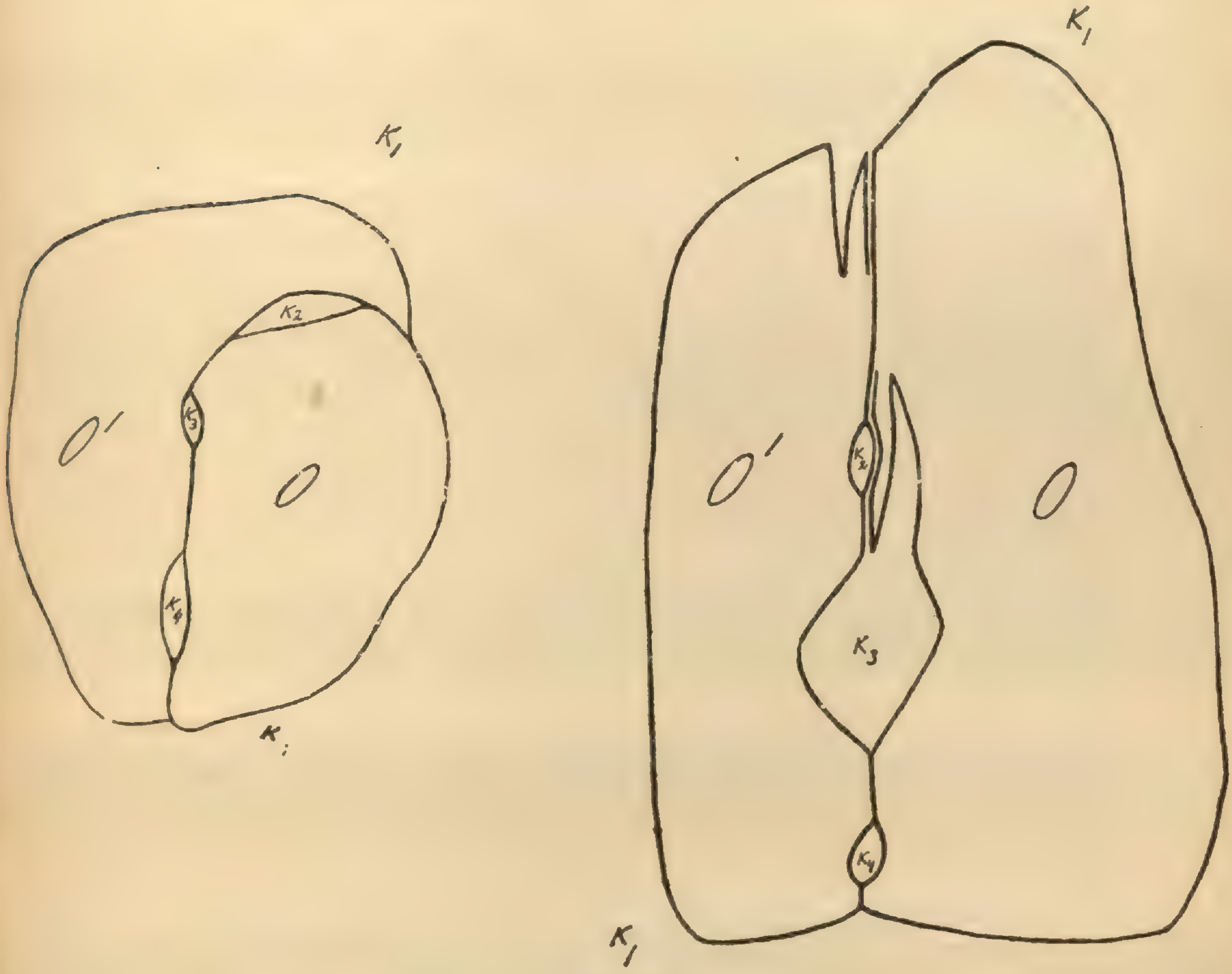

Fig. 3.

[1]) For these notions comp. L. E. J. BROUWER, "*Zur Analysis Situs*", and "*Ueber ein-eindeutige stetige Transformationen von Flächen in sich*", Mathem. Annalen, Bd. 68.

junctures [1]) Z (lying in u) and Z' (lying in u') form together a coherent set determining *one* rest region, or a point P of u, and its image P' in u' will find themselves on the same $\varkappa_n$ in such a way, that P lies outside K', and P' outside K. Then however in those points P and P' an enlargement of O and O' would be possible, which would clash with the property of a transformation domain. So we are sure, that both meetings take place in the first-mentioned way.

Let us discern in a juncture Z, performing such a meeting, its *right end* Z_r and its *left end* Z_l. Let us represent by K_r resp. K_l the branch of u approaching to Z_r resp. Z_l; by ζ_r resp. ζ_l the part of the *circumference* [2]) of Z_r resp. Z_l, approximated by K_r resp. K_l; and by ζ the complete circumference of Z. Let us further indicate $m(Z, Z')$ by T, the circumference of T by τ, (Z_r, Z'_l) by T_r, $m(Z_l, Z'_r)$ by T_l, and the part of the circumference of T_r resp. T_l, approximated by K_r and K'_l resp. K_l and K'_r, by τ_r resp. τ_l.

We at once see, that of the sets T_r and T_l at least one is coherent. For in the opposite case we could choose on K_r in the vicinity of Z_r a point of u outside K', whose image on u' would lie outside K, and an enlargement of O and O' would be possible there (see the schematic fig. 4).

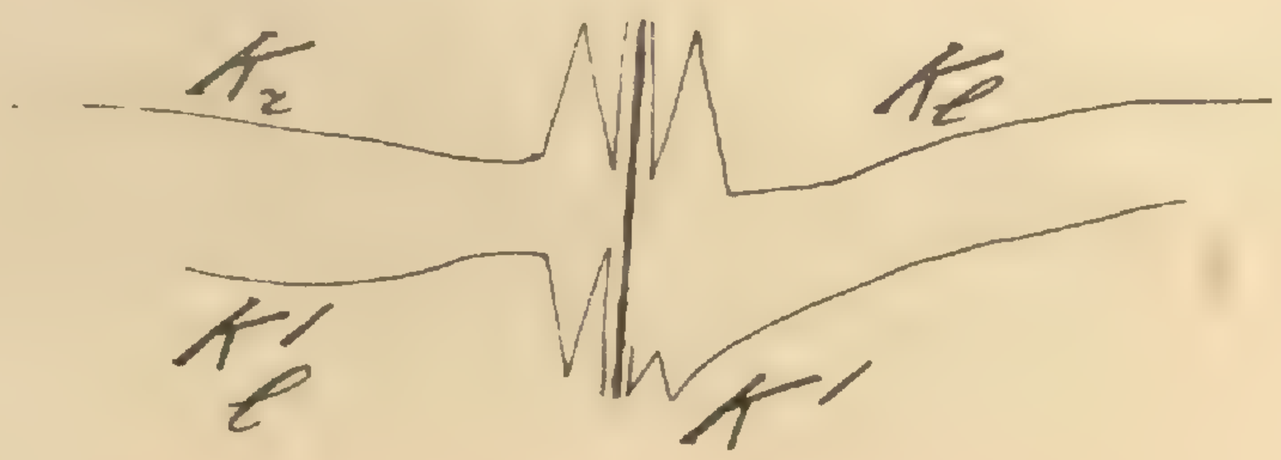

Fig. 4.

In the same way it is evident, that *either* ζ_r and ζ'_l or ζ'_r and ζ_l must be identical to each other, *or* Z must be a part of Z' or Z' a part of Z.

Otherwise namely *either* ζ_r would have to lie partly outside ζ'_l and at the same time ζ'_r partly outside ζ_l *or* ζ_l partly outside ζ'_r, and at the same time ζ'_l partly outside ζ_r, which free segments of circumference would partly correspond to each other, and would admit an enlargement of O and O' in their vicinity.

Of the two possibilities obtained we shall first discuss:

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[1]) Two arcs of curve cohere to a new arc of curve by means of a "*juncture*", which contains an *end* of each of them. In the following reasoning we suppose the considered juncture to be composed of two non-singular ends. For singular ends it needs a slight modification.

[2]) i. e. the cyclically ordered set of its accessible points.

I. ζ_r *and* ζ'_l *are identical to* τ_r. We then further notice, that ζ_l and ζ'_r cannot leave a part s of the circumference of T_r free between K_l and K'_r (so that T_l also is coherent, though of τ_l this is not yet certain).

Otherwise namely s would be a part of the circumference of T_r, outside τ_r, ζ_l and ζ'_r; if we then regard T_r as Z_r, there will correspond to s a part s' of the circumference of Z'_r outside ζ'_r, ζ_l and τ_r. So about s and s' the domains O and O' could be enlarged.

We now distinguish the following two cases:

I*a*. τ_r *and* ζ_l *do not cohere on* τ [1]); then τ_r and ζ'_r do not cohere either. We can then represent Z schematically according to fig. 5, where each of the segments represents an arc of curve, to which further ramifications may be attached; however no ramifications cohere with the schema between *ap* and *ah*, and neither between *bq* and *bk*. Furthermore we notice that in *a* as well as in *b ab* unites itself with each of the two other arcs of curve ending there to a new arc of curve. In an analogous way the sets Z' and T are represented in this figure [2]).

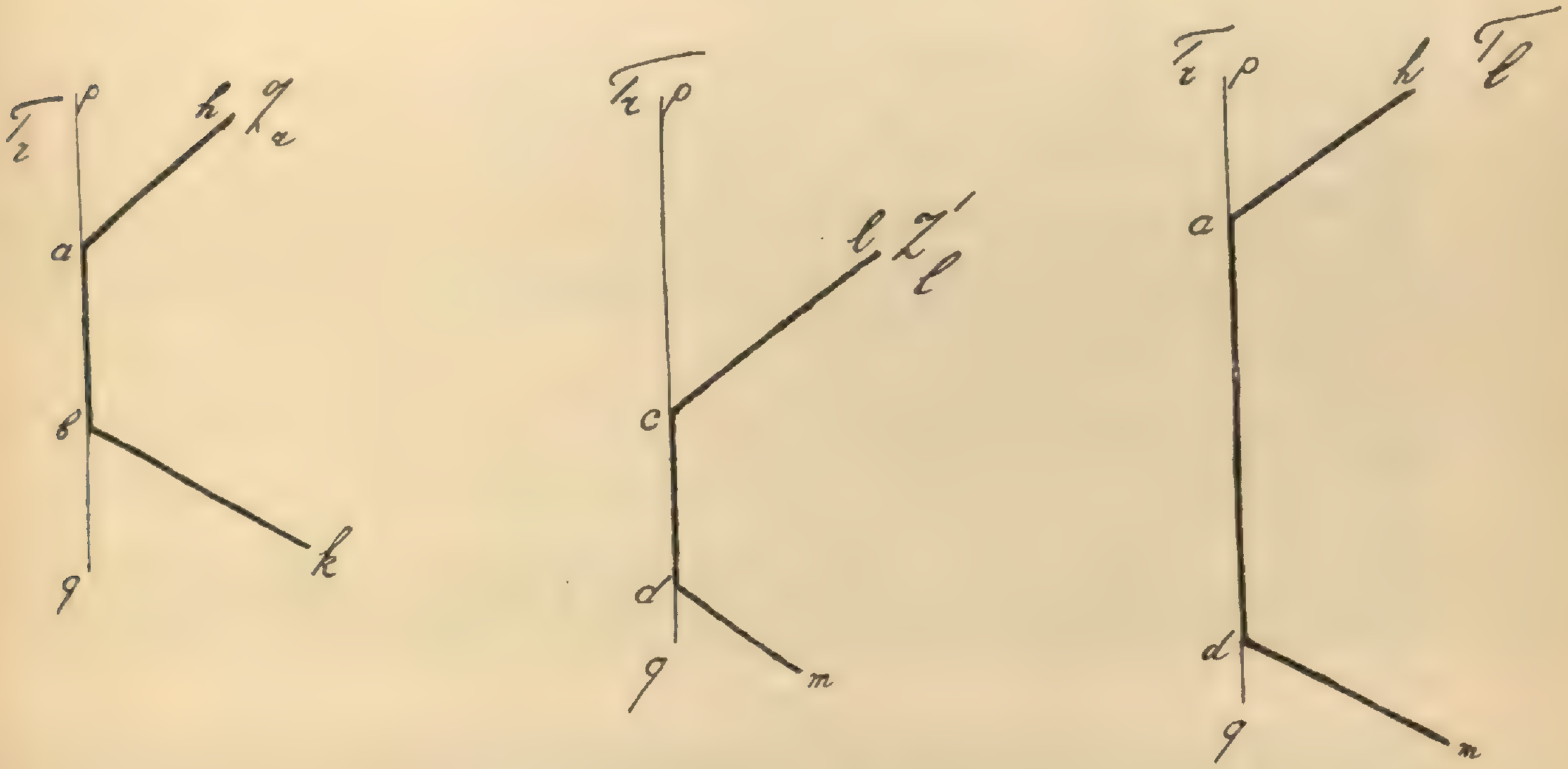

Fig. 5.

Now the situation of the three sets T_r, Z'_r, and Z_l with respect to each other can be in different ways in this case; the two essential possibilities are represented in fig. 6.

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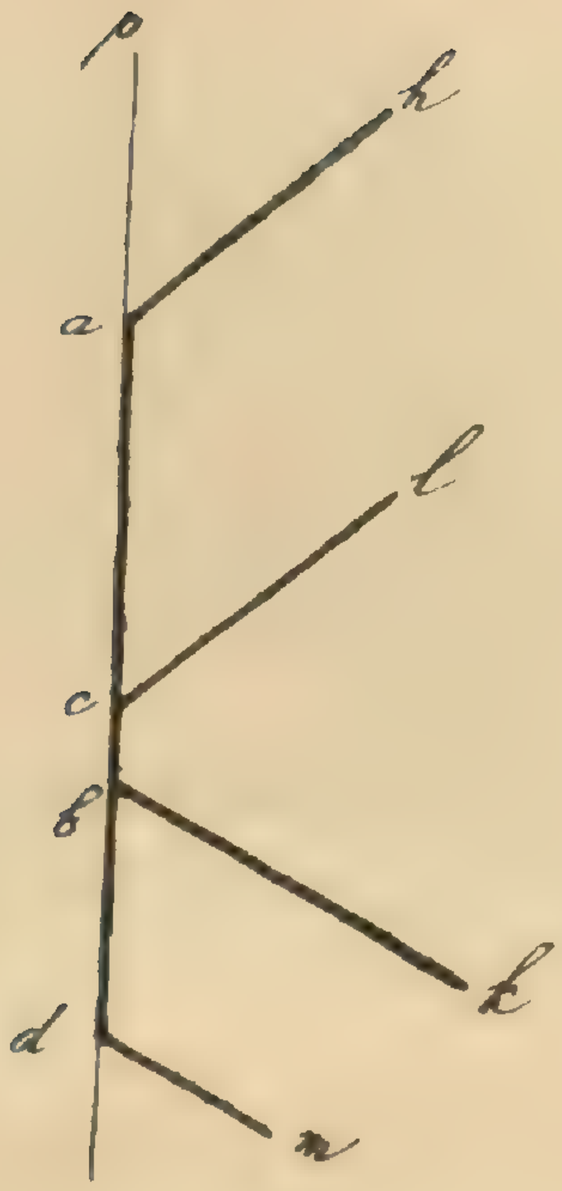
[1]) I*a* may also be treated in a less direct way, analogously to I*b*.

[2]) Some of the segments of these figures may shrink to zero; this however does not injure the correctness of our conclusions.



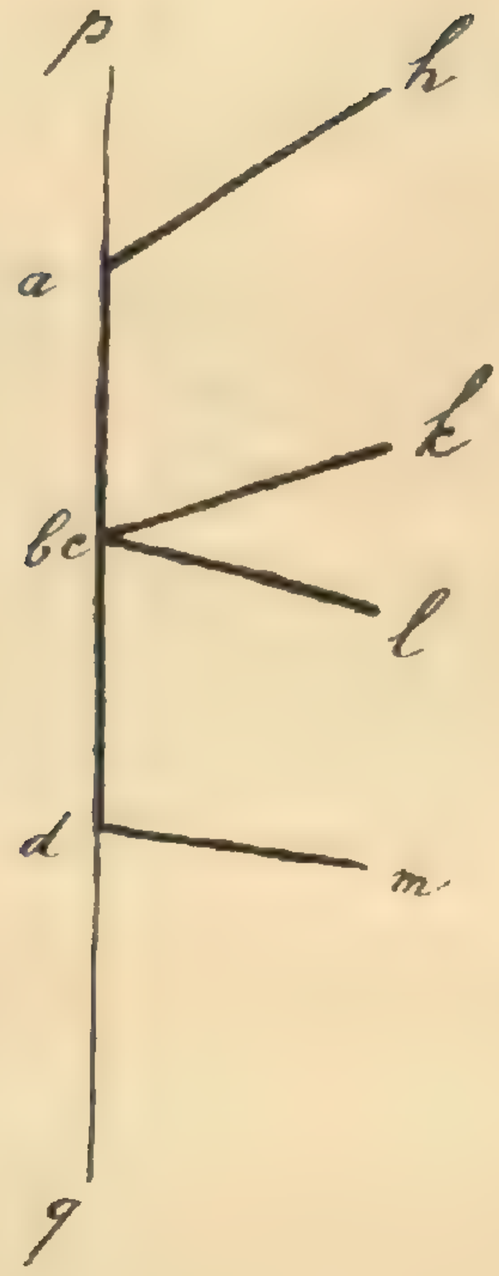


Fig. 6.

I$a\alpha$. When the first possibility of this figure appears, it follows from the correspondence of the arcs of curve ab and dc for the transformation, that on the arc of curve bc must lie somewhere a *juncture invariant for the transformation.*

I$a\beta$. When the second possibility appears, we notice that in bc the arcs of curve dc and bk certainly cohere. For otherwise ab and cl would cohere there neither, and from both would follow, that in bc between ζ'_r and ζ_l a segment s of the circumference of T_r would be free of both ζ'_r and ζ_l, the impossibility of which we have proved.

Let us furthermore in bc call L_{dc}, L_{lc}, and L_{ab} the ends of the arcs of curve dc, lc, and ab, then, $habq$ having for the transformation as its image $qdcl$, and L_{lc} and L_{ab} cohering with each other, the situation of L_{dc}, L_{lc} and L_{ab} with respect to each other is quite the same, as that of τ_r, ζ'_r and ζ_l; the former thus allow of quite the same investigation as the latter, in which they are contained. If by this investigation we arrive again at the case I$a\beta$, we can investigate still further partial sets which are in the same case, and can go on in this way. After a denumerable number of repetitions of this process we must then either have reduced this case to an other, or have found *a point invariant for the transformation.*

Ib. τ_r *and* ζ_l *partly cover each other, or at least cohere on* τ; then the same holds for τ_r and ζ'_r.

Then, as τ_r is the image of ζ_l, and ζ'_r the image of τ_r, we can,

starting at ζ_l, divide the part of τ, approximated by K' and K'', into successive "transformation domains", separated by Schnitte in this circumference, so that in a circuit each following domain is the image of the preceding one; of the last domain of the series then in general only a part appears, but three at least must be complete; fartheron the derived set of this last partial domain must cohere with that of the first domain; and we may suppose the number of the domains to be finite, as otherwise we should at once fall back on case II.

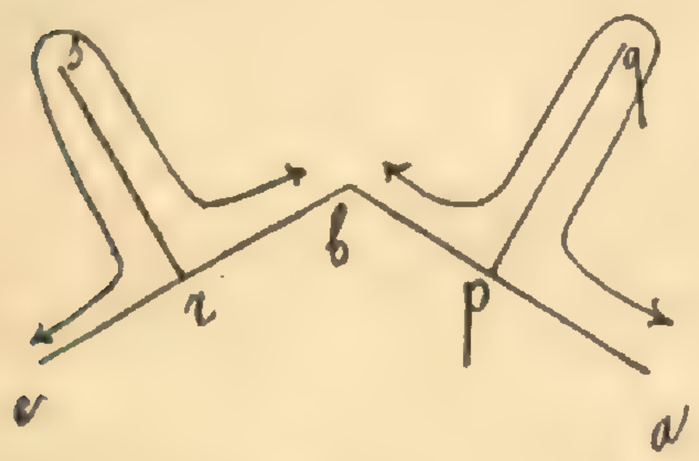


Fig. 7.

These domains of the circumference we can reduce by destroying in each of them the subsets of T, belonging to no other part of τ. If e.g. in fig. 7 the first domain extends from a to b, the second from b to c, we can deprive the first of the arc pq, the second of the arc rs.

On the circumference of the rest set R_1 we have then still a division into domains with the same properties, as the original division of the circumference of T. By a displacement of the separating Schnitte between the domains (after which in general the first as well as the last domain will be a partial one) it may occur, that the same process of reduction is once more applicable to R_1, and in this way it is repeated, until after a denumerable number of reductions a set R_0 is left, which by no displacement of the separating Schnitte can be made fit for further reduction.

A domain d then spreads over a part of the circumference h of an arc of curve k in such a manner, that the domain itself as well as the rest of the circumference possesses the whole k as its derived set.

Let the next domain d' follow of that rest of the circumference of k a part e', before it leaves k, and let f be the part of h belonging neither to d, nor to e'. Let e' be the image of e, and let us call g the segment of h approximated by k'. These notations are illustrated by two examples in fig. 8.

We now distinguish the following two cases:

I$b\alpha$. *f does not lie everywhere dense on k.* Then e must lie everywhere dense on k.

For in the opposite case we might take the part of h enclosed between the end Schnitt of e and the end Schnitt of e' as a domain, which would be capable of reduction, and this is impossible.

But if e lies everywhere dense on k, the image of k is a part of k, so that we fall back on case II.

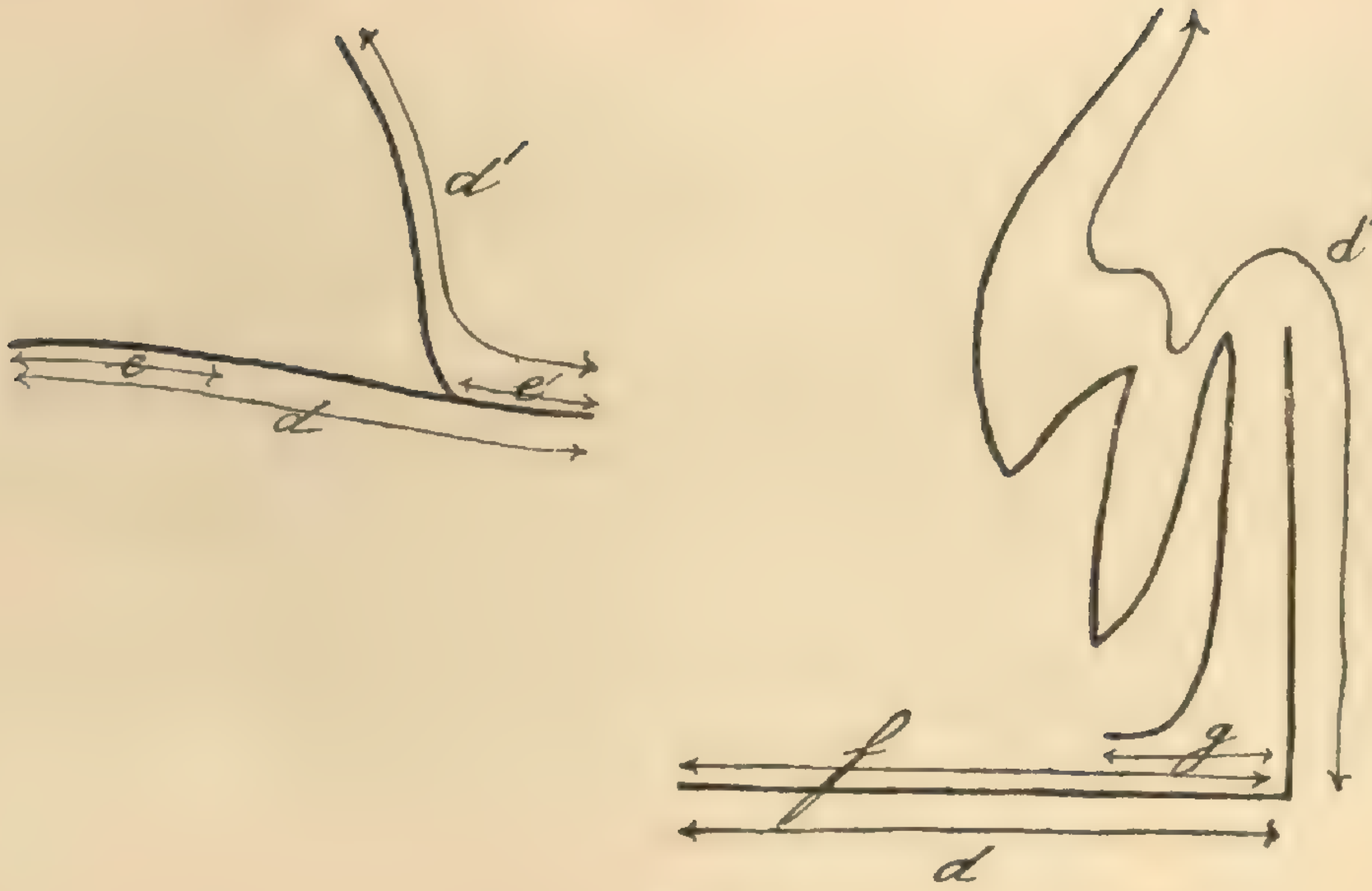


Fig. 8.

Ib β. *f lies everywhere dense on k.* Then g must also lie everywhere dense on k.

For in the opposite case the arcs of curve k, k', k'', etc. form an arc of curve B, in whose circumference the domains d, d', d'', etc. leave free a part everywhere dense in B, whilst each arc $k^{(n)}$ coheres with $k^{(n-1)}$ and $k^{(n+1)}$, but with none of the other. This however is impossible, as we have shown above, that the last and the first domain must cohere with each other.

But if g lies everywhere dense on k, k is a part of k', and again we are in case II.

II. *Z' is a part of Z.* We then, and also in the preceding cases reduced to this, have an *open system of curves*[1]) S, having as its image S' a part of itself. The image S'' of S' is again a part of S', etc. If $S^{(\infty)}$ is the set common to all sets $S^{(n)}$, it is *an open system of curves i, invariant for the transformation.*

From the commencement of the domain construction according to fig. 2 is evident, that the two sets T, in which u and u' meet each other, and therefore also the two invariant sets i, lie isolated from each other.

At the commencement of the domain construction we have presupposed, that the two arcs of curve of fig. 1 have no endpoint in common. If that *is* the case, the method needs but slight modifications, giving no difficulties. We then start not with common arcs of simple curve accessible for both O and O', but only with common points accessible for both O and O', which is sufficient too.

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[1]) This means a perfect, coherent, nowhere dense set, determining in a region of the connection of the inner region of a circle or parabola only *one* rest region.

Furthermore we have supposed in our reasoning, that K_r etc. have nothing more in common with T, than Z_r etc. However it might be possible, that the last part of e. g. the branch of K_r approximating Z_r is contained in Z'. One can easily be convinced, that this pecularity does not harm the proof.

We can sum up our results as follows:

THEOREM 1. *An arbitrary continuous one-one transformation of a twosided surface in itself with invariant indicatrix possesses a transformation domain, which either breaks the connection of the surface, or joins two isolated open systems of curves, invariant for the transformation.*

As furthermore such an invariant open system of curves possesses at least *one* invariant point, as I shall prove in another communication, the following holds likewise:

THEOREM 2. *An arbitrary continuous one-one transformation of a twosided surface in itself with invariant indicatrix possesses a transformation domain, which either breaks the connection of the surface, or joins two points invariant for the transformation.*

The fundamental importance of these theorems for the theory of transformations and transformation groups I shall show lateron. Here I only wish to indicate, how the theorem of the invariant point of the sphere, proved in my first communication upon this subject, is contained in them, and how this theorem can be extended to the Cartesian plane.

For, if the transformation domain of a sphere or Cartesian plane has more than two rest regions, then surely one of them must be invariant together with all the points of its boundary.

If we thus exclude this case, a transformation domain O on the sphere, which breaks its connection, is either annular, or singly connected; fig. 9 shows either of these possibilities.

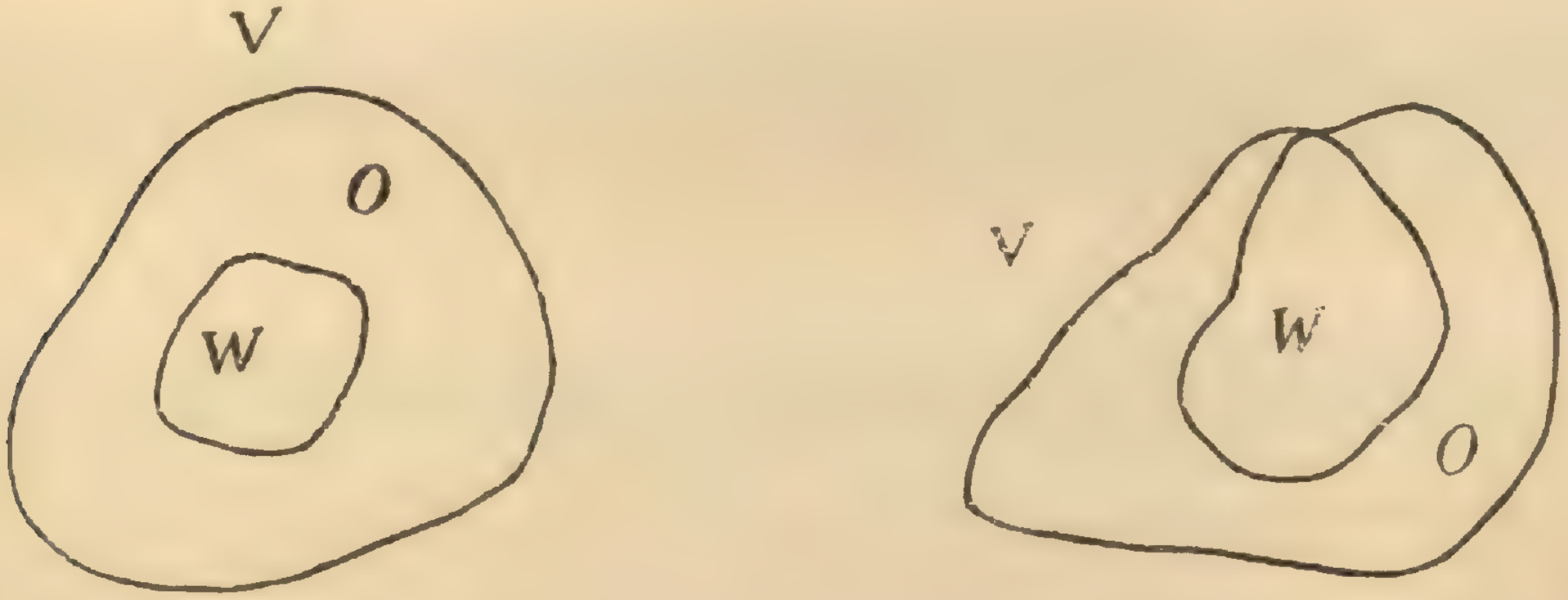

Fig. 9.

In both cases O determines on the sphere two regions, V and W, in one of which lies its image O'; then in the other lies O'_i, i.e. its image for the inverse transformation. If we now repeat the transformation itself as well as its inverse an indefinite number of times, we obtain on one hand in V a series of domains $O', O'', O''', \ldots$, and on the other hand in W a series of domains $O'_i, O''_i, O'''_i, \ldots$

In neither of these series exist intermediary regions between the successive terms, and each series converges to an invariant limit set, which causes the existence of at least one invariant point. These invariant limit sets may cohere with each other (in the second case of fig. 9); then we are sure of only *one* invariant point, otherwise always of *two*.

If in the Cartesian plane we have a transformation domain breaking its connection, either one of the cases of fig. 9 appears again, or the domain is bounded by an arc of curve running from infinite to infinite, or by two such arcs of curve.

If it is bounded by one arc, the existence of an invariant open system of curves can be deduced according to the proof of theorem 1.

If it is bounded by two such arcs B_1 and B_2, we can, if no

Fig. 10

invariant point exists, always arrange that these arcs do not cohere, and within O (see fig. 10) an arc of simple curve β can be constructed, running from infinite to infinite, lying entirely outside its image β', and enclosing with β' a new transformation domain ω. According to the SCHÖNFLIES process of representation [1]) the domains ω', ω'', ω''', ... , determined by a series of successive repetitions of the transformation, can then be represented on regions enclosed by straight lines $x = na$ and $x = (n+1)\,a$ in such a way that a series of corresponding points in $\omega, \omega', \omega'', \omega'''$,... answers to a series of points (p,q), $(p+a,q)$, $(p+2a,q)$, $(p+3a,q)$,... ; furthermore, if no invariant point exists, we can arrange, that the just-mentioned series of images of ω, continued indefinitely on both sides, covers the whole Cartesian plane, i. o. w. we have proved:

THEOREM 3. *An arbitrary continuous one-one transformation of the Cartesian plane in itself with invariant indicatrix either leaves at least one point invariant, or is a continuous one-one image of a translation.*

ERRATUM.

In my paper: "*The force field of the non-Euclidean spaces with positive curvature*", these *Proceedings* IX 1, p. 261, l. 6 from top:

for: The unilateral elliptic Sp_n is enclosed by a plane Sp_{n-1},

read: The elliptic Sp_n is enclosed by a unilateral plane Sp_{n-1}.

Chemistry. — "*On the nitration of diethylaniline*". By Prof. P. VAN ROMBURGH.

(Communicated in the meeting of September 25, 1909).

A considerable time ago I communicated to the Academy the results of the action of nitric acid on dimethylaniline dissolved in a large excess of sulphuric acid [2]). The difference in behaviour which dimethyl- and diethylaniline exhibit when nitrated under other conditions induced me to study also the action of nitric acid on diethylaniline in presence of an excess of sulphuric acid, and indeed this amine and some of its nitro-derivatives appeared to behave in many respects differently from dimethylaniline.

If we dissolve diethylaniline in double its volume of concentrated sulphuric acid and pour this solution into an excess of nitric acid

[1]) Mathem. Ann. 62, p. 319—324

[2]) Verslagen Akad. v. W. Febr. 23, 1895 and These Proc. Dec. 30, 1900.

(D. 1.48) we obtain 1.2.4 dinitrodiaethylaniline (m.p. 80°) as I have demonstrated previously [1]).

If, however, diethylaniline (1 mol.) is dissolved in twenty times its weight of sulphuric acid (D. 1.84) and if to this solution are cautiously added 2 mols. of nitric acid dissolved in about five times the weight of sulphuric acid, care being taken that the temperature of the nitrating mixture does not rise above 5°, and if, after a few hours it is poured into iced water, a more or less deep yellow, crystalline product is obtained.

If this is collected and the filtrate neutralised to a large extent with sodium carbonate, a fine dark red compound is separated.

It may be stated here that this experiment has been carried out many times; strange to say the results were often quite different although the conditions of working were practically the same; as will now appear, it depends on the quantity of iced water into which the mixture is poured.

Recrystallisation of the yellow crystalline mass from alcohol gave as main product a compound melting at 59°, a small quantity of a fine orange red product m.p. 95°, being also occasionally deposited. If the latter was recrystallised I often obtained yellow as well as orange red crystals which, however, melted at the same temperature (95°). Sometimes, on recrystallising the compound melting at 59°, the orange crystals melting at 95° were obtained, so that it looked as if the compound with the lower melting point passed into that with the higher one.

The dark red substance separated by means of the alkali, when recrystallised from alcohol, melted at 76°.

The elementary analysis gave the same result for all these substances; the figures obtained showed that they all had the composition of dinitrodiethylaniline.

The peculiar behaviour of the above products melting at 59° and 95° rendered a closer investigation desirable and finally I succeeded in finding the key to the problem; the substance melting at 59° is a compound of two different dinitroderivatives of diethylaniline, and the one melting at 95° occurs in two, and probably more, modifications.

If we dissolve one part of the substance melting at 59° in 11.4 parts of hydrochloric acid (D. 1.14) and then cautiously add water whilst shaking, a yellow product separates, which, however, soon turns orange, as a rule. After 6 parts of water have been added the crystals are collected and washed with a little hydrochloric acid

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[1]) Rec. T. II p. 36 [1883].

D. 1.08. If now to the filtrate is added much water a yellow product is precipitated. The melting point of the orange-red product is 95°, that of the yellow one 80°. This last one proved identical with the 1.2.4 dinitrodiethylaniline described previously.

In a quantitative experiment there was obtained from 0.750 gram of the substance melting at 59°, 0.370 gram of the orange and 0.358 gram of the yellow product, and from the neutralised mother liquors another 0.021 gram of solid matter was obtained.

If we dissolve a mixture of equal weights of the two nitro-compounds in boiling alcohol, the compound melting at 59° crystallises on cooling. The curve of the melting points of mixtures of both nitro-compounds in different proportions showed undoubtedly the existence of a compound formed from equal molecules of the said substances.

The compound melting at 95° is, as will be shown presently, 1.3.4- dinitrodiethylaniline. It is soluble in most of the organic solvents at the boiling heat. If the hot concentrated solution is cooled rapidly yellow crystals only are often obtained, whereas on cooling slowly orange crystals are generally formed. The yellow ones represent the labile (β), the orange ones the stable α-modification [1]). Occasionally and under definite conditions, a third modification is formed which is also yellow, but very unstable, much more so than the β-modification, and which in the dry state passes very slowly into the orange one.

The products formed in the nitration in presence of strong sulphuric acid being now known, the progressive change of the reaction was investigated more closely.

In the first place it appeared that in the case of diethylaniline, the nitration under the conditions described, that is when 2 mols. of nitric acid are used, does also not proceed further than the formation of mononitroderivatives. In fact, if to the mixture of diethylaniline with sulphuric and nitric acid, is added another mol. of diethylaniline, also dissolved in 20 parts of sulphuric acid, and the mass poured into iced water, no *di*nitro-compounds appear to have formed, and we obtain only m. and p. nitrodiethylaniline.

If, however, we dissolve m. nitrodiethylaniline in 75 times its weight of dilute sulphuric acid (1 : 2) and then add 1 mol. of colourless nitric acid, the solution obtained remains clear for days. If, however, we add a trace of sodium nitrite, a separation of the

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[1]) Prof. JAEGER has examined this compound crystallographically, also the compounds melting at 59° and 76°, for which I again wish to express my thanks. The results are described in Zeitschr. f. Krystallographie (1905) XL 129.

dinitroderivative m. p. 95° starts immediately, evidently owing to the catalytic action of the nitrous acid, and on addition of sodium carbonate to the filtrate the red product m. p. 76° is obtained [1]). p. Nitrodiethylaniline is also readily converted into the 1.2.4-dinitroderivative. We may, therefore, conclude that even when we use pure sulphuric acid and colourless nitric acid, traces of nitrous acid are formed in the nitration of diethylaniline, which accelerate the further nitration in the aqueous solution, (after pouring into iced water) particularly if the liquid has not become too diluted.

A judicious choice of the acid-concentration in the liquid obtained, after pouring the nitration mixture into water, might render it possible not to obtain the compound of 1.2.4- and 1.3.4-dinitrodiethylaniline (m. p. 59°), but these components separately, because, as we have noticed, the 1.2.4-compound, m. p. 80°, has somewhat stronger basic properties than the 1.3.4-compound.

A series of experiments now demonstrated that both conditions may be fulfilled, while it was shown also that within definite limits, a quantity of nitric acid larger than that calculated for 2 mols. has a favourable influence.

The following *modus operandi* can now be recommended when large quantities of those new dinitroderivatives have to be procured in a short time.

100 grams of diethylaniline are dissolved in sulphuric acid (D. 1.84) up to 1200 c.c. Of this solution are taken portions of 120 c.c., and after cooling to 0° a solution of 14 grams of nitric acid, (D. 1.49), in 42 grams of sulphuric acid is added, care being taken that the rise of temperature during the nitration does not exceed 5°. After the mixture has been standing for a few hours it is poured with thorough stirring into a mixture of 250 grams of water and 250 grams of ice. The final temperature is then about 30°.

The solution often clear at first soon becomes turbid and deposits the product m. p. 95°. After about an hour the liquid is filtered, and if the filtrate no longer gets turbid to any extent an equal volume of water is added whilst stirring. A yellow product is precipitated which is also collected; this is the 1.2.4-dinitrodiethylaniline melting at 80°, which is sometimes contaminated with a little red 1.3.6-dinitrodiethylaniline, particularly if more water has been used.

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[1]) If we dissolve m. nitrodiethylaniline in dilute nitric acid (D 1.22) we obtain, after a long time, besides the products melting at 95° and 76°, also a dinitromonoethylaniline 1.3.6, which is also formed in the oxidation of the product m. p. 76 with chromic acid in acetic acid solution.

On neutralisation of the filtrate with sodium carbonate the 1.3.6-derivative m. p. 76° is obtained.

We now have a complete control over the reaction. If we use more than the amount of acid indicated, we run the risk of obtaining also trinitroderivatives, which impede the purification of the 1-3-4-dinitrodiethylaniline.

When treated with nitric acid D. 1.37, the compound melting at 95° passes readily into trinitroderivatives. For this purpose 2.5 grams are dissolved in about 21 c.c. of that acid at the ordinary temperature. After standing for about half an hour yellow crystals are deposited, with evolution of heat, which are collected immediately. Generally a little nitrous acid is formed.

When carrying out this experiment for the first time, I obtained as main product a substance melting at 158° [1]). On repeating the same in order to prepare a larger quantity and using more nitric acid I obtained, however, as main product a compound melting at 175°.

Judging from the analytical results, the first one is a trinitrodiethylaniline and the other a trinitromono-compound.

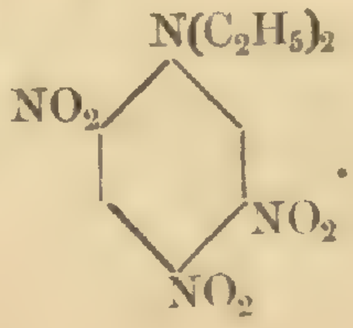
The cause of the different course of the reaction was soon found out: in the second nitration nitric acid had been used prepared by diluting yellow 90% acid, whereas, in the first one the acid D 1.37 was obtained by diluting colourless acid D 1.40 with water. Owing to the nitrous acid present in the yellow acid, one ethyl-group would have been replaced by hydrogen.

That this explanation is the correct one could be demonstrated by dissolving trinitrodiethylaniline m. p. 158° in moderately strong sulphuric acid, and adding to the solution obtained sodium nitrite. On pouring the mass into water the derivative melting at 175° separated.

This was obtained also by oxidation of the diethyl compound with chromic anhydride in acetic acid solution.

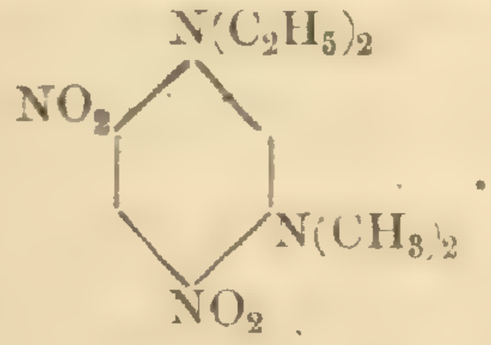
We have succeeded also in determining or deducing the structure of the compounds described.

From analogy with the results of the nitration of dimethylaniline it may be expected that the structure of the trinitro-compound melting at 158° would be:

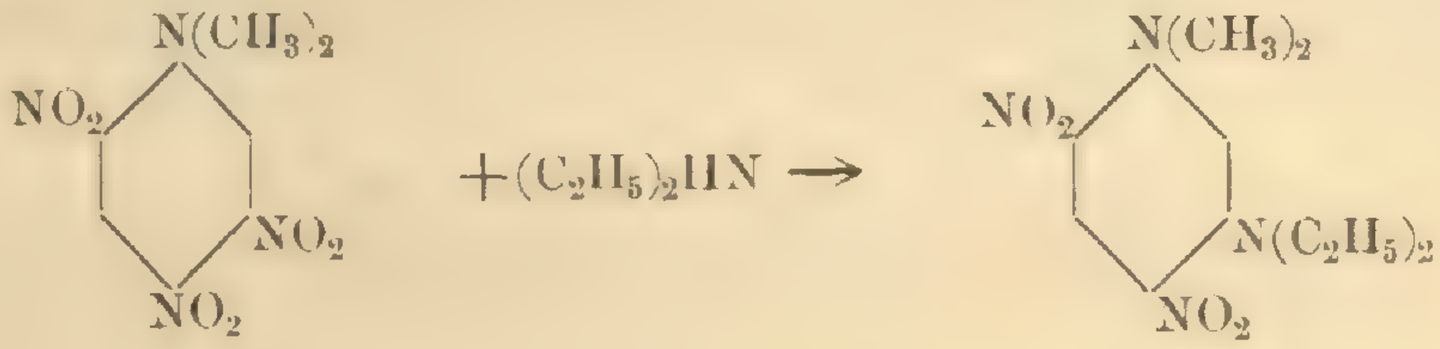


[1]) There is also formed a small quantity of an isomeride melting at 117°. By repeated crystallisations from carbontetrachloride the melting point rose to 132°.

If this hypothesis is correct a metaphenylene derivative should be formed by the action of *dimethylamine*, namely 4.6. dinitrodimethyldiethylmetaphenylenediamine :

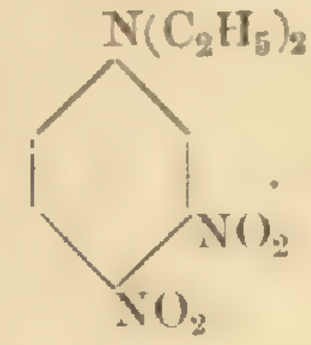


This substance must then, however, be identical with the reaction product of *diethylamine* on the 1.3.4.6. trinitrodimethylaniline m. p. 195° which I prepared previously :



Dr. Ultée was kind enough to scrutinise these reactions, and obtained indeed with both an identical product melting at 83°, and which, on analysis, gave the results required for a dinitrodimethyldiethylphenylenediamine.

A melting point determination of a mixture of the products obtained in both reactions did not show a depression of the melting point.

As regards the two new dinitro-products we must assign to the one melting at 95° the structure formula:  . One of the nitro-groups may be readily substituted by the action of ammonia or amines [1]), which points to the fact that the two nitro-groups are in the *ortho*position in regard to each other. Derivatives of *m*-phenylenediamine are then formed.

This dinitroderivative is also formed from *m*-nitrodiethylaniline whereas the formation of the 1-3-4-6-trinitroderivative m.p. 158° on subsequent nitration, agrees entirely with that view.

On the other hand, the dinitrodiethylaniline m. p. 76° does not contain any moveable nitro-group so that, taking into consideration that in the nitration with *dilute* nitric acid one nitro-group occupies

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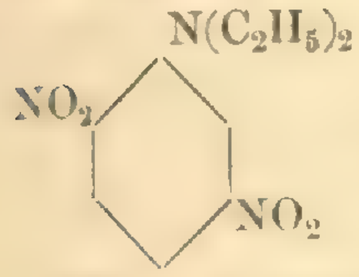
[1]) With ammonia a compound melting at 135° is formed, which in sulphuric acid solution gives a fine red coloration with traces of nitrous acid.

The description of this compound and of many others obtained in this research will appear in the "Rec.".

in regard to the amino-group the *ortho*- or *meta*-position, the structure:



should be assigned to this compound.

The structure of both these compounds is therefore quite in agreement with those of the nitro-products which I obtained previously from dimethylaniline.

Utrecht. Org. Chem. Lab. University.

Chemistry. — "*On sodium alkyl carbonates.*" By Prof. A. P. N. FRANCHIMONT.

(Communicated in the meeting of September 25, 1909).

Of alkyl carbonates but very little is known. After DUMAS and PÉLIGOT had prepared in 1840 barium-methyl carbonate and potassium-ethyl carbonate, and noticed their property of being decomposed by water into alcohol and a hydrocarbonate, BEILSTEIN gave in 1859 a good method for the preparation of sodium-ethyl carbonate. He passed dry carbon dioxide through a solution of sodium ethoxide in absolute alcohol. The sodium salt was obtained in 1868 by GEUTHER from diethyl carbonate and sodium ethoxide, the potassium salt by HABERMANN in 1886 in the electrolysis of anhydrous potassium acetate in absolute alcohol, the barium salt by DESTREM in 1882 by passing dry carbon dioxide into a solution of barium ethoxide in absolute alcohol. Attention was always called to the decomposition of these salts by water. This property is generally believed to be common to all the alkyl carbonates.

Sodium-phenyl carbonate is also decomposed by water into phenol and sodium hydrogen carbonate. Of this, however, it is also known, that it evolves carbon dioxide on warming, and this is not mentioned of the alkyl carbonates. Although, as a rule phenol and primary aliphatic alcohols have but little in common, I thought it would be advisable to make a comparative study of the alkyl carbonates and sodium-phenyl carbonate, particularly since Dr. MOLL VAN CHARANTE had found that the latter salt is decomposed at the ordinary temperature by dry acetone, with evolution of carbon dioxide and formation of phenol.

I, therefore, prepared sodium ethyl carbonate by BEILSTEIN's method, and observed that it is decomposed by water into sodium hydrogen carbonate and alcohol. It is, however, not decomposed by acetone nor when heated to about 180°.

Sodium phenyl carbonate is now prepared by the dry process, namely, by passing dry carbon dioxide over dry sodium phenate, whereas ethyl carbonate is prepared by the moist process. Dr. MOLL VAN CHARANTE having made unsuccessful experiments to prepare sodium phenyl carbonate by the moist process, I tried to prepare sodium ethyl carbonate by the dry method, in order to avoid the objection that the difference in properties between sodium phenyl carbonate and sodium ethyl carbonate might be due to the different method of preparation.

I succeeded in finding the conditions in which dry, alcohol-free sodium ethoxide gives with dry carbon dioxide a *product* which agrees in composition and properties with that prepared according to BEILSTEIN's method.

It did not yield carbon dioxide on heating, or on treatment with acetone, but was decomposed by water into alcohol and acid carbonate.

The different behaviour of sodium phenyl- and sodium ethyl-carbonate is, therefore, not due to the different manner of formation, but to the difference between alkyl and phenyl, or to a difference in the action of carbon dioxide on sodium phenyl- and ethyl-carbonate. These experiments, which I had made three years ago, have been recently repeated and extended at my instigation by Dr. DUBSKY who prepared also in both ways sodium methyl carbonate which hitherto had never been made.

Not only were all my results with sodium ethyl carbonate confirmed, but it was also shown that sodium methyl carbonate behaves entirely like its homologue towards water, acetone and heat. As, however, it was shown during the research that the acetone should be dried by shaking with phosphorus pentoxide and subsequent distillation, which was not done in MOLL VAN CHARANTE's experiments, we have repeated with his co-operation the experiment with sodium phenyl carbonate and acetone, and although we dare not pretend with absolute certainty that sodium phenyl carbonate is not affected by dry acetone, a powerful decomposition with evolution of carbon dioxide, as noticed by him, was not observed by us.

In the case of sodium methyl- and ethyl carbonate the *product*, after having been treated with acetone, was analysed and investigated and showed no difference from the original.

Chemistry. — "*On the abnormal reduction of an aromatic nitro-compound with tin and hydrochloric acid and an interesting case of dimorphism*". By Dr. Otto de Vries. (Communicated by Prof. A. P. N. Franchimont).

(Communicated in the meeting of September 25, 1909).

In the synthesis of isocreosole, one of the methyl esters of homo-pyrocatechol, 3-amino- 4-methoxytoluene had to be prepared by reduction of the corresponding nitro-compound. It appeared that this reduction proceeds very neatly and quantitatively when carried out with iron and acetic acid; if, however, tin and hydrochloric acid is used, a mixture of amino-compounds is formed from which a second, chlorinated, substance could be isolated in addition to the normal product of the reaction.

The formation of amino-compounds chlorinated in the nucleus, in the reduction of aromatic nitro-compounds with tin and hydrochloric acid, has been observed several times, and it was, therefore, interesting to determine which place the chlorine atom had occupied in this case.

This could be ascertained by substituting the amino-group by chlorine. The dichloro- 4-methoxytoluene formed appeared to be identical with 3-6-dichloro- 4-methoxytoluene, which had been prepared synthetically by substituting in Limpach's 3-amino- 4-methoxy- 6-nitro-toluene first the amino-group in 3 by chlorine, then the nitro-group in 6 by NH_2 and subsequently also by chlorine. From this it appears that, as might be expected from analogous cases, the chlorine atom had occupied the para-position in regard to the amino-group.

The 3-6-dichloro- 4-methoxytoluene just mentioned exhibited a peculiar case of dimorphism. In the various preparations, partly described, it was always obtained in the form of needles melting at 29°, which always behaved as a perfectly stable substance. At the close of the investigations, however, another form occurred, namely, flat crystals melting at 44° and from that moment, the needles melting at 29° were no longer stable; they fell to powder and then exhibited the melting point 44°. The labile form (29°) may be easily regenerated by cooling a fused mass, or by recrystallisation from dilute alcohol. After the appearance of the stable form (44°) it was, at first, impossible to isolate the needles so obtained, for as soon as they were exposed to the air of the laboratory, conversion set in. Even the precautionary measures taken by Liebermann and Bhilmann in their investigations of the cinnamic acids proved inadequate. Only by heating the substance for some length of time above 44°

in a sealed tube, disinfecting this completely by immersing it in alcohol and carrying on the further manipulations in another laboratory, the needles melting at 29° could be isolated. Afterwards, when the form (44°) had not been used for some weeks, the infection seemed to have practically vanished, and, with the usual precautions, the needles melting at 29° were stable in the air.

The conversion of the two forms into each other is readily accomplished; the fused mass yields, on cooling, the form melting at 29° which by inoculation or in an infected region passes spontaneously into the stable form 44° with generation of heat.

From this it is quite evident that we are dealing with a case of dimorphism and more in particular with a case of monotropism where the one form is always labile towards the other.

This research will be communicated in detail in the "Recueil des travaux chimiques".

Botany. — "*Contribution to the knowledge of watersecretion in plants.*" By Dr. W. Burck.

(Communicated in the meeting of September 25, 1909.)

In a previous paper "*On the biological significance of the secretion of nectar in the flower*" [1]) I pointed out the correspondence between the secretion of nectar in the flowering period and of water or mucilaginous fluid in the closed flower-bud. On continuing the investigation there arose a doubt in my mind as to the truth of prevailing views on water-secretion at the surface of the plant.

This induced me to make some observations, the results of which will be communicated here, which give a different view of what is to be understood by water-secretion.

I propose to show later that these modified views on water-secretion are not without significance for our conception of floral and extrafloral nectar-secretion.

The phenomenon that in many herbaceous plants and shrubs drops of water are secreted during the night and the early morning hours at the tips and margins of the leaves (guttation) is ascribed to the power of the roots for forcing up considerable quantities of water under favourable conditions; when the air cools down and approaches the dew-point this water is forced out in the form of

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[1]) These Proceedings, Nov. 28, 1908.

drops. At the place where the drops appear there are generally found peculiar pores; these water-stomata were first distinguished by DE BARY from ordinary stomata.

It is assumed that the secretion of water is of importance to the plant in order to prevent the danger, that water, forced up by root-pressure, should become subject to increased pressure in the vascular bundles in consequence of impaired transpiration, and should thus be injected into the intercellular spaces.

Of late years doubt has occasionally arisen whether in all plants and under all circumstances this guttation is only to be ascribed to the forcing up of the water taken up from the soil by the roots, and whether it is to be regarded as filtration pure and simple.

In 1895 HABERLANDT [1]) expressed the view that in some plants the anatomical structure of the leaf-tooth and especially of the tissue between the terminal tracheids of the vascular bundle and the water-pores — the so called epithema (DE BARY) — suggested a glandular function. He also showed experimentally that in *Conocephalus ovatus*, for instance, this tissue really plays a part in the secretion of water. In *Fuchsia*, however, where the structure of the epithema also very strongly suggests a gland, the process was nevertheless found to depend in filtration, but here the tissue was supposed to keep the system of intercellular spaces between vascular bundles and water-cavities filled with water in order to ensure the separation of the tracheal system.

In any case HABERLANDT considered the glandular action to be dependent on root-pressure: the gland was only supposed to act when it was stimulated by the increased pressure of bleeding.

An investigation by NESTLER [2]) in 1896 clearly showed, however, that the phenomenon does not always depend on root-pressure.

NESTLER found that cut leafy shoots of *Tropaeolum majus* which were kept under a moist bell-jar with their cut surface in water, showed after three days drops of water on the younger leaves, at the termination of the vascular bundles; i. e. in the same place where under ordinary circumstances the phenomenon of guttation occurs; on the fourth day the formation of drops was also observable on some older leaves.

NESTLER remarks in this connexion, that here the epithema cannot be concerned in the secretion: in the first place this tissue is not

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[1]) G. HABERLANDT, Ueber wassersecernirende und absorbirende Organe. Sitzungsberichte der Kais. Akad der Wissensch. in Wien. Abth. II Vol. 104, 1895.

[2]) NESTLER, Sitzungsberichte der Kais. Akad. der Wissensch. Wien. Vol. 105. 1896. p. 524; Vol. 106. 1897. p. 401. Vol. 108. 1899, p. 690.

well defined in *Tropaeolum* and secondly drops of water also appear on the stem, where there can be no question whatsoever of epithema. The secretion there takes place through water-pores which in position and shape differ more or less from the ordinary stomata. He believes that the explanation of the phenomenon must be sought in osmotic forces, which are called into play under the circumstances mentioned, and cause the secretion.

Properly speaking this was not the first time that this phenomenon was mentioned.

As early as 1881 WILSON [1]) believed he had observed formation of drops on the cut branches of *Impatiensglandulifera*, *I. parviflora* and *Fuchsia*, but an investigation of GARDINER [2]) soon showed that, as regards *Impatiens*, there could be no question in WILSON's experiments of the secretion of water, but of nectar, whereas in *Fuchsia* the fluid observed by WILSON was not derived from the water-pores of the teeth, but was secreted by water-glands in the neighbourhood of these pores.

It is very remarkable that the phenomenon here in question has not been observed much earlier, for, as I have found, there are undoubtedly numerous plants which, under the above-mentioned external conditions, quite independently of root-pressure or artificial pressure, show the formation of drops in the clearest manner. I have observed this formation for instance in *Kerria japonica*, *Philadelphus coronarius*, *Weigelia rosea variegata*, various species of *Spiraea*, *Hydrangea pubescens*, *Hydrangea campanulata*, *Deutzia crenata*, *Cornus sibirica* and in other species of *Cornus* and I am convinced that afterwards, in a more detailed treatment of this subject, I shall have no difficulty in showing it is the case with many other plants.

In all these plants clear drops appear after some hours on cut branches under the bell-jar at the tip and margin of young leaves; on removal the drops are renewed.

It will be shown that these drops are formed in definite leaves, and that they owe their origin in some cases to mucilage-glands which later only secrete water, and in other cases to glands which secreted water from the beginning.

We may safely assume that in the case of many plants in which

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[1]) W. P. WILSON, On the cause of the excretion of water on the surface of nectaries. p. 11. Leipzig 1881.

[2]) W. GARDINER, On the physiological significance of waterglands and nectaries. Proceedings of the Cambridge Philosoph. Society. Vol. V, p. 34—50.

one sees in their natural condition at night or in the morning the formation of a certain number of drops of water on the leaves, a part of these drops have passed out independently of the direct influence of root-pressure.

The phenomenon of guttation may be regarded as the formation, under certain external conditions, of drops of water at the tip and the margin of the leaf, whether from the water-pores as a direct consequence of root-pressure, or as a result of the activity of internally situated glands. Further investigation of water-secretion has shown me, however, that this phenomenon of guttation only gives a very incomplete idea of what water-secretion in the plant may be considered to be.

There are numerous plants which do not show proper drop-formation, but which nevertheless continually secrete at their surface fairly considerable quantities of water or watery mucilage — especially in their younger parts.

It may be remembered that TREUB [1]) showed more than 20 years ago, that there is another kind of water-secretion in plants, by epidermal trichomes.

TREUB found that the flower-buds of a tropical Bignoniacea, *Spathodea campanulata* always contain water which is most probably secreted from the cushion-shaped trichomes, which cover the inside of the calyx in large numbers.

Afterwards, as is well known, the same phenomenon i. e. secretion of water in a closed flowerbud, was observed in other tropical plants; in these cases similar trichomes were always observed, whether only on the inner surface of the calyx or also on the outer surface of the corolla, and there was every reason to assume that the fluid was secreted by these epidermal trichomes.

For a long time this phenomenon was considered a thing apart. There was no inducement to connect this secretion of water inside the flower with that outside on the leaves. It was regarded as a special contrivance, a useful adaptation, brought about by natural selection in order to protect the young floral parts inside the calyx against too strong transpiration.

A few years after TREUB's paper, HABERLANDT, however, discovered that such a water-secretion, resulting from the secretory action of externally situated glands, can also be observed outside the flower.

During his stay at Buitenzorg he found on the surface of the leaves

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[1]) TREUB M. Annales du Jardin botanique de Buitenzorg. Vol. VIII. 1890.

of *Spathodea campanulata* the same trichomes which secrete the calyx-water inside the flower and he could show experimentally that under favourable conditions these latter are also able to secrete water.

In addition HABERLANDT described in other plants metamorphosed epidermal cells and multicellular trichomes on the surface of the leaves, which under certain conditions functioned as water-glands.

HABERLANDT's observation regarding the occurrence of water-glands on the leaves was soon followed by others.

NESTLER gave a detailed account of the water-secretion from epidermal glands in *Phaseolus multiflorus* and in Malvaceae and VON MINDEN [1]) in *Nicotiana* and *Glaux maritima*.

With regard to one very important point, viz. the conditions under which water-secretion from the glands takes place, HABERLANDT, however, reached a conclusion which is not in agreement with the results of earlier observers, particularly with those of GARDINER on the water-secretion from the multicellular epidermal glands on the leaf surface of *Limoniastrum (Statice) monopetalum* and from the water-glands of Polypodiaceae: nor has HABERLANDT's conclusion been confirmed by the above-mentioned later investigations.

Starting from the view that the secretion of water would serve to prevent an overloading of the leaf with water and an injection of the intercellular spaces, HABERLANDT regarded the glands on the surface of leaves and also those of ferns and similar glands as *regulators of the water-content* which only begin to act when the hydrostatic pressure in the vascular-system, i.e. the so-called bleeding-pressure, has reached a certain height through interrupted transpiration.

NESTLER [2]), to whose investigations on water-secretion in leaves of *Phaseolus multiflorus* we owe the certain knowledge, that the water is really excreted by the glands (he succeeded in directly observing under the microscope the excretion of drops of water from the cells of the glandular hairs), was unable to confirm this view [3]). He found that the secretion of water is independent of root-pressure, as GARDINER had also found for *Limoniastrum* and for ferns. The drops of water appeared on the surface of cut leaves which were kept in a space saturated with aqueous vapour.

[1]) M. VON MINDEN, Beiträge zur anatomischen und physiologischen Kenntnis wassersecernirender Organe. Bibliotheca botanica Heft 46. 1899.

[2]) l. c. Bd. CVIII. 1899 und Berichte der deutsch. bot. Gesellsch. Bd. XVII 1899. S. 332.

[3]) That we cannot always deduce from the presence of glands that any water which may occur has been secreted by those glands was shown by NESTLER's investigation of water-secretion in *Boehmeria* (Sitzungsber. Wien Vol. CVIII, p. 690).

Von Minden obtained the same result from his experiments. He found that the secretion of water from the glands on the leaves of *Nicotiana* and of *Glaux maritima* took place equally well with cut branches placed under a bell-jar, with the cut surface in water, as with root-bearing plants. He suggested that many of Haberlandt's plants might have behaved in the same way, in as much as the glands of *Phaseolus multiflorus* correspond closely to these of *Nicotiana*, and those of *Bignonia brasiliensis*, *Spathodea campanulata*, species of *Piperomia*, *Artocarpus integrifolia* correspond to the glands of *Glaux maritima*.

It will be shown below that this suggestion was perfectly legitimate. In my own investigations I have found so many plants which have epidermal glands which are independent of root-pressure that I do not hesitate to express the conviction that this is the general rule.

Investigation has also shown me, however, that such water-glands which secrete a more or less mucilaginous fluid, are much more common in plants than has hitherto been supposed — especially in the younger parts — and that in arriving at a conception of the process of water-secretion we must in the first place take into account these ever-functioning epidermal glands.

Thus I have observed a secretion of water on the leaves of: *Philadelphus coronarius*, *Hydrangea pubescens*, *Hydrangea campanulata*, *Weigelia rosea*, *Deutzia crenata*, *Corylus Avellana*, *Ulmus campestris*, *Sambucus nigra*, *Syringa vulgaris*, *Forsythia viridissima*, *Fuchsia* spec., *Calystegia sepium*, *Datura Stramonium*, *Nicandra physaloides*, *Cosmos hybridus*, *Dahlia variabilis*, *Melandrium album*, *Thymus Serpyllum*, *Malva silvestris*, *Sidalcea candida*, *Abutilon Darwinii*, *Abutilon Avicennae*, most of which will be further considered below.

First, however, it may be as well to point out that we may deduce from Reinke's paper in Pringsheim's Jahrbücher of 1876 [1] on the histological structure of so-called secretion-organs, occurring on leaves, that Reinke had already observed that the exudation of drops of water at the apex or the teeth of the margin of leaves may be a result of glandular activity.

Reinke's paper is a continuation of Hanstein's investigation of resin- and mucilage-secreting structures in buds, which was published

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[1] Reinke, J., Beiträge zur Anatomie der an Laubblättern, besonders an den Zähnen derselben vorkommenden Secretionsorgane. Pringsheim's Jahrb. Vol. X. 1876 p. 119.

in the Botanische Zeitung of 1868 [1]). In this paper HANSTEIN had pointed out the fact, then almost unknown, that in the first stages of the development of buds in many dicotyledonous plants, trichomes of the most different forms occur on the very young leaves but especially on the stipules and leaf-sheaths; sometimes these trichomes consist of bristles or woolly hairs, often also of colleters, which possess the power of secreting a mixture of resin (or balsam) with mucilage, the so-called Blastocolla; by this sticky substance the youngest leaves are surrounded.

REINKE now showed that this secretion in the bud of mucilagenous and resinous substances, which HANSTEIN had brought to light, is much more general in plants than HANSTEIN had led one to expect, and that these glands especially occur at the teeth of the leaf-margin. In his paper he gives a description of the structure of the glandular teeth in no less than 83 species, belonging to widely different orders, and here and there he also indicates how the secretion-product is extruded.

REINKE summarized his results as follows:

"Die auf vorstehenden Blättern gegebenen Mittheilungen zeigen, "dass die Sägezähne am Rande der Dicotylen-Blätter in den meisten "Fällen Träger eigenthümlicher Organe sind, von denen sich in der "Regel eine secernirende Thätigkeit nachweisen lässt, die entweder "in der Zeit des Knospenzustandes oder in spätere Altersstufen der "Blätter fällt . . . ."

"Was das gelieferte Secret anlangt, so ist dasselbe in der Knospe "ein zäher flüssiger Schleim oder Harz, am ausgebildeten Blatte nur "eine wässerige oder etwas schleimige Flüssigkeit; im Einzelnen sind "darüber besondere Untersuchungen anzustellen."

"Was die verschiedene Form der Drüsenorgane der Blattzähne "anlangt, so gruppiren sich dieselben naturgemäss in äussere hervor- "tretende und eingesenkte Drüsen; die ersteren bilden eine stufen- "weise sich ändernde Reihe, die folgende Typen umfasst: *Kerria*, "*Prunus*, *Betula*, *Corylus*, wo das secernirende Organ im ersten Fall "aus einem ganzen Blatt-Abschnitt, im letzten Falle nur aus einem "aufgesetzten Trichom besteht. Die zweite Hauptform, ... ist besonders "zahlreich und tritt erst am entwickelten Blatte deutlich hervor, um "hier unter günstigen Umständen klare Tropfen auszuscheiden. Wenn "die hierher gehörigen Zähne auch bereits in der Knospe Schleim "secerniren, so verhalten sie sich mit *Kerria* übereinstimmend."

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[1]) HANSTEIN, Prof. J., Über die Organe der Harz- und Schleim-Absonderung in den Laubknospen. Bot. Zeit. 1868. p. 697, 721, 745, 756.

"Combinirt treten beide Formen z. B. bei *Viola* auf," u. s. w.

In order to make this quotation more clear I add that REINKE found in *Kerria japonica* that the portion of the leaf-margin situated above the termination of the vascular bundle — therefore that portion where in many plants a so-called epithema is found — consists of a mucilage-secreting gland. In *Prunus*, as long as the leaf is young, each tooth of the leaf bears a dark red conical projection, which secretes balsam abundantly during the bud-stage. In *Betula* a large number of peltate glands (Trichomzotten), are found on the surface of the young leaves and internodes, while the leaf-teeth terminate in an egg-shaped mucilage gland. The latter only secretes in the young bud, whereas the peltate glands seem to function principally in the fully-developed leaf. In *Corylus* the young leaves are also covered over the whole of their surface with glands (Trichomzotten) which have the form of pedunculated buds. In addition every leaf-tooth also terminates in such a gland. The latter secretes in the young bud; the others are only developed afterwards.

It may be deduced from this quotation that according to REINKE a phenomenon, comparable to guttation, may arise in various ways, independently of root- or bleeding-pressure, as a result of the activity of glands. This activity may be due to internally situated glands of the leaf-margin, or to secretion of water by externally situated trichomes which at an earlier stage secreted mucilage (resin, balsam) or finally it may be the result of both internal and external glands.

Except for the fact that the secretion of water is not always preceded by that of mucilage and that conversely the secretion of mucilage is often not followed by that of water, I have confirmed REINKE's results.

To the circumstance that in discussing his results REINKE made only a passing reference to secretion of water and did not refer to it at all in the body of his paper, nor mentioned experiments relating thereto, must be attributed the fact that this reference did not receive much attention, although his paper became widely known.

I will now mention some observations which show that secretion of water in the plant can indeed be brought about by various ways.

As will be seen, the problem demands a more detailed and systematic treatment; I therefore intend to revert to it at a later time.

In the first part of this communication I will limit myself to the secretion of water in leaves, and return afterwards to that in flowers.

I. *Secretion of water in leaves.*

Kerria japonica.

The leaf-apex and the teeth of the leaf-margin are acuminate and lanceolate. The vein running into the tooth ends at some distance from the apex; above this there are a number of water-pores resembling stomata.

The portion of the leaf-tooth above the termination of the vascular bundle, where in other plants there is a so-called epithemal tissue of peculiar structure which has more or less the properties of spongy parenchyma, consists in *Kerria japonica* of a mucilage-gland of which both the epidermal and the interior cells are rounded and filled with highly refractive substances. The secretion of mucilage occurs in young-leaves in the bud; it takes place with elevation of the cuticle.

Similar glands but somewhat larger and more rounded are also found at the apex and margin of the young stipules and especially also at the edge of the calyx-segments; they differ from the glands of the leaf-margin in shape and in size, but the cells of which they are composed have the same highly refractive contents.

If a cut branch of this plant is placed in a moist space a formation of drops soon takes place at the apex and the teeth of the young leaves, as was mentioned above. The drops secreted from the same cells which formerly, in the bud, secreted mucilage, contain no mucilage or at most very little. If these drops are removed with a strip of blotting-paper, new drops soon make their appearance.

The same phenomenon occurs in the young flowerbud. The water is secreted in the form of droplets which flow together to a few large drops. There can be no question of a secretion of water by means of an epithema. It must undoubtedly depend on the endosmotic activity of the gland-cells which with great force draw water from the surrounding tissues and from the vascular bundle and press it out.

Philadelphus coronarius.

In the second half of April when the buds began to open, a cut branch was placed in a glass of water under a glass bell-jar which was kept moist.

The outermost leaves of the bud, which before had their margins adjoining, began to separate. The next pair of leaves, placed alternately, thus became visible; of this pair the margins still adhered. The remaining leaf-pairs were still enclosed.

Already on the evening of the same day, clear droplets could be seen at the apex and the teeth of the outermost leaves, and at the apices of the next pair of leaves, while the space between each pair of young leaves was filled with water.

The apex and the teeth of the budleaflets differ especially in anatomical structure from the rest of the leaf in having larger epidermal and subepidermal cells, which have a denser contents and contain stomata at an early age.

REINKE is of the opinion that these cells secrete mucilage in the younger stages of the bud, but I have not been able to observe this. I consider that in this case the secretion of water is not preceded by that of mucilage, but that the glandcells secrete water from the outset.

The surface of the leaf is further covered with numerous unicellular long, pointed hairs, which are laid down very early; their rounded base is afterwards surrounded by prolonged epidermal cells.

From these pointed hairs with rounded base the water is doubtless derived which under the bell-jar fills the space between each pair of leaves; they are therefore true waterglands, not glands which formerly secreted mucilage (balsam or resin).

A month later when the 4 or 5 outermost pairs of budleaves were fully grown, the experiment was repeated. The adult leaves had now lost their power of secreting water and there now only appeared waterdrops on the two pairs of small immature leaves and on a pair of leaves of the bud.

Hydrangea pubescens.

On the 1st of May a cut branch was submitted to the experiment in a glass of water under a moist glass bell-jar. The outermost pair of leaves of the buds was not yet fully grown, but nevertheless the leaves had already taken up a horizontal position. The following pair of leaves was still erect, but the leaf-margins which at an earlier stage were closely applied, began already to separate; the other leaves still formed a completely closed bud.

The leaf of *Hydrangea pubescens* terminates in a long apex and bears on its margin a large number of long narrow teeth, which point outwards in the upper half of the young leaf and which in the lower half are more or less directed backwards.

In each tooth a vein terminates and above this there are one or more waterpores. The young leaves are further covered above and below with long unicellular hairs: those of the upper surface are broad, rounded above and have very thin walls, those of the lower

surface are more pointed, have thicker walls and are marked with protuberances.

Next morning there were numerous clear drops, both at the apex and on many of the teeth, but only on those leaves which had not bent over and become free of the bud; the apices of the following third pair of leaves also showed the formation of drops.

In addition to the secretion at the teeth and at the apex, which is to be ascribed to the same cause as in *Philadelphus coronarius*, namely to the secretory action of the epidermal and sub-epidermal cells, we observe, in addition, that the hairs with which the leaf is covered, behave as glands and secrete the water with which the young leaves are covered.

In *Hydrangea campanulata* the mode of watersecretion is not very different from that in *H. pubescens*.

Weigelia rosea variegata.

In this *Weigelia* the young leaves have both halves rolled up to the middle vein. Secretion of large, clear drops of water takes place at the apex of the young leaves and at the apices of the teeth, which anatomically agree with those of *Hydrangea*. The leaf surface, especially the upper one, is thickly covered with trichomes of various sizes, shaped like a pedunculate cushion (Köpfchendrüsen), which is so often found in watercalyces; they occur especially on the basal half of the leaf They secrete abundantly so that the space enclosed by the rolled-up leaves is filled with water.

They do not occur in the very youngest leaves. The youngest leaves, of which the teeth are not yet formed, only have a covering of long unicellular hairs, of which the rounded base takes part in the secretion of water.

In an experiment made in the end of May, when the leaves of the bud were fully grown, no formation of drops took place under the circumstances mentioned.

Corylus Avellana L.

In *Corylus Avellana* every leaftooth bears at its apex a large, compound, almost cylindrical gland, by way of appendix. The teeth are placed in two rows, one of which consists of much smaller teeth which are pointed inwards; these smaller teeth also have the character of mucilage-glands.

In the branch, on which the experiment was made, the buds were just beginning to unfold. The outermost leaf had already liberated itself from the bud and was bent outwards; the glands at the apices

of the teeth and the smaller leafteeth, which are turned inwards, had at this stage already lost most of their contents and had become brown. In the younger leaves they produced much mucilage and the youngest leaflets of the bud were completely enveloped in it. In addition to these large glands every leaf bears, especially on its upper surface, a large number of smaller mucilage-glands, which too, are stalked and are compound and more or less spherical; furthermore the leaf is also covered with long thick-walled bristles. Not only the leaves, but also the stipules bear in the bud numerous mucilage-glands of both kinds: larger ones at the margin in a pretty close row between the hairs and smaller ones on the leaf surface, especially on the lower portion of the stipule. In the very youngest stipules and leaves these small spherical glands cannot yet be observed; they only appear when the large glands at the margin and at the apex have already lost their contents; as in *Kerria japonica* the apex and the margin of the leaves of *Corylus* show a growth in advance of that of the rest of the leaf.

In the moist atmosphere under the glass bell-jar the outermost leaflets, which are most advanced in development, become covered on their upper surface with numerous clear drops of fluid. Often the hairs at the edge of the leaflets are covered with clear droplets. This fluid is secreted by the glands of the leaf surface, which originally secreted mucilage, but now secrete water containing but little mucilage. On the first day there is seldom any real dripping, but the secretion of water may nevertheless be relatively important.

Ulmus campestris [1]).

In *Ulmus campestris* the leaves are folded up in the bud round the middle vein and are placed to the left and right of the central axis.

When, on the 2nd of May, a cut branch was placed under the glass bell-jar, the lowest leaf of most of the buds had unfolded itself and was beginning to turn itself outwards; the next leaf was still erect but the halves of the leaf had begun to separate a little; the higher leaves were still all folded up and enclosed by the stipules.

The young leaves bear long, pointed, unicellular hairs with rounded base, which are bent parallel to the leaf-surface; in addition these leaves are covered especially on the upper surface, with mucilage glands (Trichomzotten) which consist of 4 cells, viz. of a short basal cell, a cylindrical stalk-cell and two gland-cells placed one above the other, of which the upper one is hemi-spherical; the glands

[1]) Verg. Reinke l.c. p. 156.

produce much mucilage. The leaf teeth have two shapes; between the larger egg-shaped ones, rounded in front, there are smaller teeth turned more inwards.

As REINKE already observed a lateral vein of the leaf terminates in every leaftooth, just below its apex; above this there are some stomata. The secretion of mucilage takes place from the epidermis, the cells of which contain mucilage; the epidermis contains in addition larger cells, completely filled with mucilage. These mucilage-cells are also found in the sub-epidermal layer of the tissue. Often their number is so large, that as a result the leaftooth shows numerous transparent spots.

But besides the secretion of mucilage by the numerous teeth and glands of the leaf surface, there is also a secretion by the stipules and especially by those which are placed higher on the axis. These are covered on their inner sides with very numerous glands of the same shape as those which occur on the leaves, and they too bear numerous large mucilage-cells in their epidermis.

The youngest leaves and stipules are covered with a thick layer of mucilage.

In the moist space under the bell-jar the outermost leaves were already completely covered with fluid next morning. This fluid consisted of water or very watery mucilage and had evidently been secreted from the glands of the leaf surface, which, although originally mucilage-glands, afterwards behave as water-glands.

At that time there was no proper formation of drops, but some hours later it could be observed at the apex and the teeth.

Sambucus nigra.

The young leafteeth are completely hidden, because the leaves are rolled up very closely in the bud; they lie against the leaf surface and curve upwards. The secretion by the leafteeth is limited to the extreme apex, which resembles that of *Kerria japonica*. In the youngest bud-stages there is here a secretion of mucilage.

In addition the leaf surface is closely covered on its inside and outside with strongly secreting club-shaped trichomes, the shape, development and mucilage-secretion of which have already been described in detail by HANSTEIN (l. c. p. 731).

In the experiment with a cut branch under a bell-jar a fine formation of drops is soon observed at the apices of the young leaves, Further it becomes evident on unrolling the leaves, that the inner space is filled with water, while numerous drops of water are also found on the outside. The latter fluid is certainly derived from the

glands described by HANSTEIN, and the droplets which occur at the apices of the leaves, have here again the same origin as in *Kerria japonica*.

Syringa vulgaris.

During the bud-stage the leafapex of *Syringa vulgaris* exactly resembles that of *Kerria japonica*; the epidermal and sub-epidermal cells are in this case unusually large and they are filled with highly refractive contents. They secrete mucilage but soon degenerate. In consequence there is later no secretion of waterdrops. There are no leafteeth.

The upper and lower surface of the young leaf, especially the upper surface and the margin, are covered with numerous mucilage secreting trichomes, which have been described and figured by HANSTEIN (l.c. p. 733) and which during the bud stage secrete considerable quantities of mucilage.

If a cut branch is placed in a moist space the leaflets are soon covered by a layer of moisture. In the bud the young leaves are rolled up over one another; the intervening spaces are filled with water. The mucilage-glands (Keulenzotten of HANSTEIN) which therefore behave as water-glands, can be easily observed on the young leaves with a lense; they no longer occur on the adult leaflets.

Forsythia viridissima.

Forsythia viridissima in the main resembles *Syringa vulgaris*. The mucilage-glands have here a different form and do not mutually resemble each other.

They are generally made up of a stalk, which some times consists of one and some times of two rows of three cells, and of an almost spherical head of gland-cells proper.

Their number is if possible even larger than in *Syringa* and when there is secretion of water in a moist space there is not unfrequently formation of actual drops.

In conclusion I wish to append a note on *Fuchsia*, the plant which has first been mentioned in the literature on this subject, but which nevertheless cannot be reckoned among those plants which clearly show the phenomenon.

Fuchsia.

It has already been stated, that WILSON's observation on the water-secretion of cut branches of *Fuchsia* in a very moist space has been

confirmed to the extent that HABERLANDT noticed a formation of drops on the leaf-teeth.

I myself have, no more than NESTLER, been able to distinguish with certainty the formation of droplets on the leaf-teeth.

The latter bear at their apex a single pore with a large opening, which leads to a relatively small cavity filled with fluid, below which the epithema is situated. Of this tissue HABERLANDT has given a very detailed description and some figures.

As regards the structure of the bud, REINKE already pointed out that at the base of each leaf there are two small acicular stipules, which broaden out at the base, and secrete much mucilage with which the youngest parts of the bud are completely surrounded. Speaking generally, these are again completely simular to the tops of the leaf-teeth of *Kerria* and the cells of which they are composed, are distinguished by their highly refractive contents. In addition the lower surface of the young leaves in the bud is thickly covered with thin walled trichomes which have a broad club-shaped end and are already formed when the leaves are still extremely minute.

Here and there the same hairs occur also on the upper surface of young leaves, especially on the basal portions and they are further found in large numbers on the young flowerbuds and stalks.

For the rest the upper surface of the leaf bears somewhat longer pointed hairs with thicker walls and a cuticle with markings.

If a cut plant is placed under the moist bell-jar, the phenomenon to which GARDINER drew attention, is soon observed i. e. the upper and especially the under surfaces of the young leaves appear bedewed with numerous fine droplets.

This phenomenon is brought about by the vigorous secretion of the numerous club-shaped hairs referred to above; each of these bears at its end a droplet of water, thus showing itself to be a water-gland. This secretion may be so copious, that the lower surface of the leaf is soon (on the 2nd or 3rd day) covered with a layer of water.

The same may be observed on the young flowerbuds; these are soon surrounded by as many hundreds of small clear waterdrops as there are glands which have found a place on the surface.

It follows from the above, that in considering the secretion of water on the surface of the plant, we must not only take into account the formation of drops which result directly from root-pressure, but also

that which is brought about the apex and the margin of the leaf and on the leaf-surface as a result of the action of glands.

In many cases the glands are originally mucilage-glands (Colleteren, Keulenzotten, Trichomzotten) which secrete resin or balsam in the bud, as proved to be the case in *Kerria*, *Sambucus*, *Corylus*, *Ulmus*, *Syringa*, *Forsythia*, but in other plants they are from the beginning real water-glands: *Philadelphus*, *Deutzia*, *Hydrangea*, *Weigelia*, etc.

Physics. — "*On the theory of the* ZEEMAN-*effect in a direction inclined to the lines of force.*" By Prof. H. A. LORENTZ.

(Communicated in the meeting of June 26, 1909)

§ 1. Certain phenomena observed by HALE in sun-spot spectra have induced me to work out the theory of the ZEEMAN-effect on the assumption that the direction of observation is oblique to the lines of force, a problem that has already been treated by VOIGT [1]), but in which some details remained to be examined.

Our subject will be the "inverse" effect, to which the direct one is intimately related, and we shall start from the fundamental equations in the form I have given them in a recent article in the "Mathematische Encyklopädie" [2]), supposing the magnetic field to be homogeneous and parallel to the axis of z.

We shall assume that the particles of the body through which the light is propagated, unless they be magnetically isotropic (i. e. of such a structure that a rotation of a particle in the field has no influence on the frequency of its free vibrations) are turned by the magnetic force in such a manner that a certain "axis" proper to each particle takes the direction of the field. We shall further imagine that each particle contains a certain number of electrons forming by their arrangement some definite and regular configuration, and capable of vibrating about their positions of equilibrium under the joint influence of "quasi-elastic" forces, of resistances and of the action exerted by the external field. Though, on account of the complexity of its structure, the mode of motion of a particle may be far from simple, we can easily treat it mathematically in a general way. This is due to the circumstance that, under certain simplifying restrictions,

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[1]) W. VOIGT, Weiteres zur Theorie der magneto-optischen Wirkungen, Ann. Phys. I (1900), p. 389.

[2]) H. A. LORENTZ, Theorie der magneto-optischen Phänomene, Encyklopädie d. math. Wiss. V 22, p. 199.

the electromagnetic action exerted by a particle is found to be wholly determined by its electric moment. Therefore, in considering the influence of a particle on the propagation of light, we may replace it by a single electron, to which we may assign an arbitrarily chosen charge **e** and whose displacements **x, y, z** have such values that the products **ex, ey, ez** are equal to the components of the electric moment of the particle. This imaginary electron may be called the "equivalent electron".

If a particle were outside the magnetic field (but in the position it really has in the field) and if it were free from resistances and from the influence of the other particles, its electrons would be able to vibrate in a number of definite modes. We shall suppose that, in these circumstances, there are certain groups of "fundamental vibrations", of such a kind that all the vibrations belonging to one and the same group have a common frequency n_0, corresponding to a definite spectral line. Whenever it is necessary, we shall distinguish the different groups from each other by the indices $a, b, c, \ldots$, and we shall denote by k the number of modes of vibration in a group.

Let us next suppose the magnetic field to be excited, without, however, as yet introducing the resistances and the mutual actions between the particles. Then, instead of any group consisting of k modes of vibration with equal frequencies n_0, we shall have k modes whose frequencies are unequal, all differing slightly from this original common value. In order to distinguish these k modes, we shall assign to each of them an index $(\varkappa)$, which we shall write on the right-hand side and at the top of the symbols relating to the mode in question.

Now, in each of these fundamental vibrations that can go on in the magnetic field under the circumstances just stated, the equivalent electron will have a motion which, according to the theory of vibrating systems, must be, generally speaking, a harmonic elliptic vibration. It can be further specified, if we take into account the states of polarization observed in the ZEEMAN-effect. From these one can infer that the path of the equivalent electron must be, either a straight line in the direction of the field, or a circle whose plane is at right angles to it. The index $\varkappa_1$ will be applied to those fundamental modes for which the first case occurs, and similarly the index $\varkappa_2$ to the second case; if we want to distinguish whether the circular motion of the equivalent electron is in the direction corresponding to that of the lines of force, or in the opposite one, we shall use the index $\varkappa_{2+}$ or $\varkappa_{2-}$. However, in order not to encumber

our formulae with too many indices, we shall omit them whenever this can be done without fear of confusion.

The states of motion $\varkappa_2$ always occur in even number; in fact, corresponding to each state $\varkappa_{2+}$, there will be a state $\varkappa_{2-}$ whose frequency $n^{(\varkappa_2+)}$ lies at the same distance from n_0 as $n^{(\varkappa_2-)}$, but on the other side of it. The modes of motion $\varkappa_1$ are conjugate two by two in the same way, with the exception, however, when k is odd, of one of them, which has the original frequency n_0.

The introduction of complex expressions, after the manner generally followed in problems of this kind, will be found very convenient. By this method one finds, for each mode of vibration, definite ratios between the quantities representing the components of the displacements of the several electrons from their positions of equilibrium. These ratios determine what may be called the "forms" of the vibrations, and it is especially to be noticed that, whereas in a particle subjected to the magnetic field only, the vibrations of the k modes mentioned above have unequal frequencies, a periodic external electric force can produce forced vibrations in these different modes, all taking place with the period of the force itself.

§ 2. This case of an impressed electric force occurs when a ray of homogeneous light is propagated through the system, so that, if n is its frequency, the complex expressions for the dependent variables all contain the factor e^{int}. While a particle is vibrating, in all its different modes at the same time, under the influence of the alternating electric force $\mathfrak{E}$ existing in the beam of light, it has an electric moment whose components are

$$\mathfrak{p}_x = \mathbf{ex}, \quad \mathfrak{p}_y = \mathbf{ey}, \quad \mathfrak{p}_z = \mathbf{ez},$$

and the body is therefore the seat of an electric polarization (electric moment per unit of volume) for which we may write

$$\mathfrak{P} = N\mathfrak{p},$$

where N is the number of particles in unit of volume.

The equations of motion of a particle lead to the values [1])

$$\begin{aligned}
\mathfrak{P}_x &= Q_{2+}(\mathfrak{E}_x + i\mathfrak{E}_y) + Q_{2-}(\mathfrak{E}_x - i\mathfrak{E}_y),\\
\mathfrak{P}_y &= Q_{2+}(\mathfrak{E}_y - i\mathfrak{E}_x) + Q_{2-}(\mathfrak{E}_y + i\mathfrak{E}_x),\\
\mathfrak{P}_z &= Q_1\,\mathfrak{E}_z,
\end{aligned}$$

the coefficients Q_1, Q_{2+}, Q_{2-} indicating to what amount the vibrations in the modes $\varkappa_1$, $\varkappa_{2+}$, $\varkappa_{2-}$ contribute to the polarization $\mathfrak{P}$. The first of these coefficients is given by

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[1]) Cf. Math. Encykl. V 22, § 47.

$$Q_1 = -\underset{ab..}{S}\ \underset{\varkappa_1}{\Sigma} \frac{B^{(\varkappa)}}{\left(n + \frac{n_0^{\,2}}{n^{(\varkappa)}}\right)(n - n^{(\varkappa)}) - ing}, \quad \ldots\ldots \quad (1)$$

and for Q_{2+} and Q_{2-} we have similar expressions, containing sums which relate to the modes of motion $\varkappa_{2+}$ and $\varkappa_{2-}$. The symbols $\underset{\varkappa_1}{\Sigma}$, $\underset{\varkappa_{2+}}{\Sigma}$, $\underset{\varkappa_{2-}}{\Sigma}$ serve to indicate sums that must be extended either over the modes $\varkappa_1$, or over the modes $\varkappa_{2+}$ or $\varkappa_{2-}$ of one of the groups $a, b, \ldots$, and all these groups have their share in the sums $\underset{ab..}{S}$. The coefficient g, which we shall suppose to have the same value for all terms belonging to one and the same group, represents the influence of the resistances, and may be regarded as a measure of the breadth of the absorption lines. As to the quantities $B^{(\varkappa)}$, these are all real and positive; their value depends on the structure of the particles and is the same for two conjugate vibrations.

Let us denote by $\mathfrak{D}$ the dielectric displacement, so that

$$\mathfrak{D} = \mathfrak{E} + \mathfrak{P}, \quad \ldots\ldots\ldots \quad (2)$$

and let us abbreviate by putting

$$1 + Q_1 = S_1,$$
$$1 + Q_{2+} + Q_{2-} = S_2,$$
$$Q_{2+} - Q_{2-} = R.$$

Then we have the relations

$$\left.\begin{aligned} \mathfrak{D}_x &= S_2\,\mathfrak{E}_x + iR\,\mathfrak{E}_y, \\ \mathfrak{D}_y &= S_2\,\mathfrak{E}_y - iR\,\mathfrak{E}_x, \\ \mathfrak{D}_z &= S_1\,\mathfrak{E}_z, \end{aligned}\right\} \quad \ldots\ldots\ldots \quad (3)$$

which are to be combined with the general equations of the electromagnetic field

$$rot\ \mathfrak{H} = \frac{1}{c}\dot{\mathfrak{D}}, \quad \ldots\ldots\ldots \quad (4)$$

$$rot\ \mathfrak{E} = -\frac{1}{c}\dot{\mathfrak{H}}. \quad \ldots\ldots\ldots \quad (5)$$

In these latter formulae $\mathfrak{H}$ denotes the magnetic force that belongs to the beam of light and alternates with its frequency.

§ 3. We shall examine the propagation of plane waves in a direction lying in the plane xz, and making a positive sharp angle ϑ with the axis of z. Let the variable quantities which determine the state of the system contain the coordinates and the time only in the factor

$$e^{in\left[t-\frac{(\mu)}{c}(z\cos\vartheta+x\sin\vartheta)\right]} \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

The quantity (μ), which we shall have to determine further on, may properly be called the complex index of refraction, and if

$$(\mu)=\mu-\frac{ich}{n}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

μ will be the real index of refraction, and h the index of absorption, the amplitude diminishing in the ratio of 1 to e^{-hl} when a distance l is travelled over.

From (5) we infer

$$\left.\begin{aligned}\mathfrak{H}_x&=-(\mu)\,\mathfrak{E}_y\cos\vartheta,\\ \mathfrak{H}_y&=(\mu)\,(\mathfrak{E}_x\cos\vartheta-\mathfrak{E}_z\sin\vartheta),\\ \mathfrak{H}_z&=(\mu)\,\mathfrak{E}_y\sin\vartheta,\end{aligned}\right.$$

and then from (4)

$$\left.\begin{aligned}\mathfrak{D}_x&=(\mu)^2\,(\mathfrak{E}_x\cos\vartheta-\mathfrak{E}_z\sin\vartheta)\cos\vartheta,\\ \mathfrak{D}_y&=(\mu)^2\,\mathfrak{E}_y,\\ \mathfrak{D}_z&=-(\mu)^2\,(\mathfrak{E}_x\cos\vartheta-\mathfrak{E}_z\sin\vartheta)\sin\vartheta.\end{aligned}\right\} \quad . \quad . \quad . \quad (8)$$

If here we substitute the values (3), and if we put

$$\frac{(\mu)^2}{S_2}=1+\xi, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

$$\frac{S_1}{S_2}=1+\eta,\ \frac{R}{S_2}=\zeta, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

we get the relations

$$\left.\begin{aligned}\mathfrak{E}_x+i\zeta\,\mathfrak{E}_y&=(1+\xi)\,(\mathfrak{E}_x\cos\vartheta-\mathfrak{E}_z\sin\vartheta)\cos\vartheta,\\ \mathfrak{E}_y-i\zeta\,\mathfrak{E}_x&=(1+\xi)\,\mathfrak{E}_y,\\ (1+\eta)\,\mathfrak{E}_z&=-(1+\xi)\,(\mathfrak{E}_x\cos\vartheta-\mathfrak{E}_z\sin\vartheta)\sin\vartheta.\end{aligned}\right.$$

Before proceeding further it will be well to turn the axes of x and z in their plane over an angle ϑ, so that the second of them takes the direction of the rays. Calling the new coordinates x' and z', we have

$$\mathfrak{E}_x=\mathfrak{E}_{x'}\cos\vartheta+\mathfrak{E}_{z'}\sin\vartheta,\quad \mathfrak{E}_z=-\mathfrak{E}_{x'}\sin\vartheta+\mathfrak{E}_{z'}\cos\vartheta,$$

by which our last three equations become

$$\left.\begin{aligned}\mathfrak{E}_{z'}\sin\vartheta+i\zeta\,\mathfrak{E}_y&=\xi\,\mathfrak{E}_{x'}\cos\vartheta,\\ -i\zeta\,(\mathfrak{E}_{x'}\cos\vartheta+\mathfrak{E}_{z'}\sin\vartheta)&=\xi\,\mathfrak{E}_y,\\ (1+\eta)\,\mathfrak{E}_{z'}\cos\vartheta&=(\eta-\xi)\,\mathfrak{E}_{x'}\sin\vartheta.\end{aligned}\right.$$

Finally, if the value of $\mathfrak{E}_{z'}$ drawn from the third equation is substituted in the first and the second, we get the following relations between the transverse components of the electric force

$$\left.\begin{aligned} \{\xi(1+\eta\cos^2\vartheta)-\eta\sin^2\vartheta\}\,\mathfrak{E}_{x'} &= i\zeta(1+\eta)\,\mathfrak{E}_y\cos\vartheta, \\ -i\zeta\{(\cos^2\vartheta+\eta)-\xi\sin^2\vartheta\}\,\mathfrak{E}_{x'} &= \xi(1+\eta)\,\mathfrak{E}_y\cos\vartheta, \end{aligned}\right\} \quad . \quad (11)$$

from which we deduce by eliminating these components

$$\xi^2(1+\eta\cos^2\vartheta)-\xi(\eta-\zeta^2)\sin^2\vartheta-\zeta^2(\cos^2\vartheta+\eta)=0, \quad . \quad (12)$$

or

$$(\xi+\zeta^2)(\xi-\eta)\sin^2\vartheta+(\xi^2-\zeta^2)(1+\eta)\cos^2\vartheta=0. \quad . \quad (13)$$

If the frequencies n_0, the frequencies $n^{(x)}$ (such as they are under the influence of the external magnetic field), the resistances g and the coefficients $B^{(x)}$ are known, the quantities S_1, S_2, R and, by (10), η and ζ will be wholly determined for any chosen value of the frequency n. After having calculated the value of ξ from equation (12), we can deduce from it, first, by means of (9), the complex index of refraction (μ) and then, by means of (7), the real index of refraction μ and the index of absorption h.

Moreover, when ξ has been found, the equations (11) give the ratio between the components $\mathfrak{E}_{x'}$ and $\mathfrak{E}_y$, and also that between $\mathfrak{D}_{x'}$ and $\mathfrak{D}_y$, which has the same value, because, on account of (8),

$$\mathfrak{D}_{x'}=(\mu)^2\,\mathfrak{E}_{x'}.$$

The result is

$$\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}}=-i\,\frac{\zeta(\cos^2\vartheta-\xi\sin^2\vartheta+\eta)}{\xi(1+\eta)\cos\vartheta}\;; \quad . \quad . \quad . \quad . \quad (14)$$

it determines the state of polarization for any beam that can be propagated in the manner specified by (6), for any "principal beam", as we shall say.

Whereas the component $\mathfrak{E}_{z'}$ may very well be different from zero, the equations (8) show that $\mathfrak{D}_{z'}=0$, as might have been expected beforehand. Hence, at every point of the system, the extremity of the vector $\mathfrak{D}$ describes an ellipse in a plane perpendicular to the direction of propagation. This line, which shows us the state of polarization of the principal beam, may be called its "characteristic ellipse"; equation (14) determines, not only its shape and position, but also the direction in which it is described.

It must further be noticed that, on account of the relation (2), the ratio $\frac{\mathfrak{P}_y}{\mathfrak{P}_{x'}}$ is equal to the ratios $\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}}$ and $\frac{\mathfrak{E}_y}{\mathfrak{E}_{x'}}$, the equality of which has already been mentioned. Hence, remembering that the components of $\mathfrak{P}$ are proportional to those of the displacement of the equivalent electron in a particle, one easily sees that, while a particle is made to vibrate in its different modes of motion (in the way determined by the sums in Q_1, Q_{2+} and Q_{2-}) the projection of the equivalent electron on the wave-front moves in an ellipse of the same form

and position as the characteristic ellipse, and described in the same direction.

As (12) gives two values of ξ, there are *two* principal beams, differing from each other by their states of polarization, their velocities of propagation and their indices of absorption. All these details depend on the angle ϑ and in general on the value chosen for the frequency n.

§ 4. The magnetic components belonging to one of the members of a definite group a, b, or c etc. lie within a narrow strip of the spectrum, which we shall likewise denote by the letter a, b, or c etc. We shall confine ourselves to the propagation of light belonging to one of these parts, say to a, and we shall assume that the distances of this part from the parts b, c etc. are very great in comparison both with the breadth of a and with that of b, c etc. On this assumption the part

$$-\sum_{\varkappa_1} \frac{B^{(\varkappa)}}{\left(n+\frac{{n_0}^2}{n^{(\varkappa)}}\right)(n-n^{(\varkappa)})-ing}$$

of (1), which relates to the group b for instance, may be simplified by writing n_{0a} instead of n, n_{0b} for each $n^{(\varkappa)}$, and

$$n^2{}_{0a} - n^2{}_{0b}$$

for each denominator. The result is

$$\frac{1}{n^2{}_{0b}-n^2{}_{0a}} \sum_{\varkappa_1} B^{(\varkappa)}.$$

The quantities Q_{2+}, Q_{2-} may be treated in the same way and we can repeat for the groups c, d, .. what we have done with b. If we assume that for each group [1])

$$\sum_{\varkappa_1} B^{(\varkappa)} = \sum_{\varkappa_{2+}} B^{(\varkappa)} + \sum_{\varkappa_{2-}} B^{(\varkappa)} = 2 \sum_{\varkappa_{2+}} B^{(\varkappa)}, \quad . \quad . \quad (15)$$

the parts contributed by the groups b, c, .. taken together, to the quantities Q_1, Q_{2+}, Q_{2-} may be represented by s, $\frac{1}{2}s$, $\frac{1}{2}s$, where s is a real quantity, constant through the region a. As for the parts due to the group a, in these we may replace every denominator by

$$2\, n_0\, (n - n^{(\varkappa)}) - i\, n_0\, g,$$

understanding by n_0 the value n_{0a}. We shall simplify still further by assuming that the group a is a magnetic triplet, so that it comprises but one mode of vibration $\varkappa_1$, one mode $\varkappa_{2+}$ and one $\varkappa_{2-}$. The frequencies of the free vibrations in these modes are

$$n_1 = n_0, \quad n_{2+} = n_0 + v, \quad n_{2-} = n_0 - v, \quad . \quad . \quad . \quad (16)$$

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[1]) See § 51 of the Article in the Math. Encykl. cited above.

where v has a value proportional to the strength of the field. Moreover, if the relation (15) is supposed also to hold for the group a, we may put

$$B^{(\varkappa_{2+})} = B^{(\varkappa_{2-})} = \alpha n_0 \quad , \quad B^{(\varkappa_1)} = 2\,\alpha n_0 \quad ,$$

with a positive constant α, so that we find

$$Q_1 = s - \frac{2\alpha}{2(n-n_1)-ig} \, ,$$

$$Q_{2+} = \tfrac{1}{2}\, s - \frac{\alpha}{2(n-n_{2+})-ig} \, ,$$

$$Q_{2-} = \tfrac{1}{2}\, s - \frac{\alpha}{2(n-n_{2-})-ig} \, .$$

By this the values of S_1, S_2 R, η and ζ have likewise become known. It is easily seen that $1 + s = {\mu_0}^2$, when μ_0 is the real index of refraction that would be found for $n = n_0$ if the particles were not put in motion in the modes of the group a, but only in those of the groups b, c, . . . If, finally, we put

$$\alpha = {\mu_0}^2 \beta \, ,$$

$$u_1 = \frac{\beta}{2(n-n_1)-ig} = \beta \frac{2(n-n_1)+ig}{4(n-n_1)^2+g^2} \, , \quad . \quad . \quad . \quad . \quad . \quad (17)$$

$$u_{2+} = \beta \frac{2(n-n_{2+})+ig}{4(n-n_{2+})^2+g^2} \quad , \quad u_{2-} = \beta \frac{2(n-n_{2-})+ig}{4(n-n_{2-})^2+g^2} \, , \quad . \quad . \quad (18)$$

we get

$$\eta = \frac{u_{2+}+u_{2-}-2u_1}{1-(u_{2+}+u_{2-})}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (19)$$

$$\zeta = \frac{u_{2-}-u_{2+}}{1-(u_{2+}+u_{2-})}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (20)$$

and, after having calculated ξ by means of (12),

$$(\mu)^2 = {\mu_0}^2 \{1 - (u_{2+} + u_{2-})\}(1 + \xi) \quad . \quad . \quad . \quad . \quad (21)$$

In the large majority of cases the absorption, even at the place in the spectrum where it is strongest, is very feeble along a distance of a wave-length. Consequently, the quantities u are very much smaller than 1. Equations (19) and (20) show that η, ζ are very small, and by (12) ξ is so likewise, so that (19), (20) and (21) may be written [1])

$$\eta = u_{2+} + u_{2-} - 2\,u_1,$$

$$\zeta = u_{2-} - u_{2+} \, ,$$

$$(\mu) = \mu_0 \left\{1 - \frac{1}{2}(u_{2+} + u_{2-}) + \frac{1}{2}\,\xi\right\} . \quad . \quad . \quad . \quad (22)$$

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[1]) Many of VOIGT's equations are free from these approximations. See also § 11 below.

§ 5. By putting $\vartheta = 0$ or $\frac{1}{2}\pi$, we are led back to the well known theory of the ZEEMAN-effect for directions parallel or perpendicular to the lines of force. Indeed, from (13) we deduce for the first case

$$\xi = \pm \zeta \, . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (23)$$

and for the second

$$\xi = -\zeta^2 \text{ or } \xi = \eta \, .$$

Further, when $\vartheta = 0$, we have by (14)

$$\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}} = \mp i,$$

so that in this case, whatever be the value of n, one of the principal beams, corresponding to the upper sign, is characterized by a left-handed, and the other by a right-handed circular polarization. This will require no further explanation. We may, however, say some words about the rotation of the plane of polarization that is observed along the lines of force, and especially about its amount for $n = n_0$. In this case

$$u_1 = i\frac{\beta}{g}, \; u_{2+} = \beta\,\frac{-2\,v + i\,g}{4\,v^2 + g^2}, \; u_{2-} = \beta\frac{2\,v + i\,g}{4\,v^2 + g^2}, \quad . \quad . \quad (24)$$

$$\zeta = \frac{4\,\beta\,v}{4\,v^2 + g^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (25)$$

Hence, according to the formulae (23) and (22), the complex index of refraction is

$$(\mu_+) = \mu_0 \left\{1 - i\,\frac{\beta\,g}{4\,v^2 + g^2} + \frac{2\,\beta\,v}{4\,v^2 + g^2}\right\}$$

for the left-handed beam, and

$$(\mu_-) = \mu_0 \left\{1 - i\,\frac{\beta\,g}{4\,v^2 + g^2} - \frac{2\,\beta\,v}{4\,v^2 + g^2}\right\}$$

for the right-handed one.

Comparing these expressions with (7), we see that the two rays are equally absorbed, the index of absorption being for both of them

$$h = \frac{n_0\,\mu_0\,\beta\,g}{c\,(4\,v^2 + g^2)}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (26)$$

but that their real indices of refraction are unequal. Their difference is given by .

$$\mu_+ - \mu_- = \mu_0\,\frac{4\,\beta\,v}{4\,v^2 + g^2},$$

and, corresponding to it, there is a rotation of the plane of polarization amounting to

$$\psi = \frac{n_0}{2c}(\mu_- - \mu_+) = -\frac{n_0\,\mu_0}{c}\frac{2\,\beta\,v}{4\,v^2 + g^2} \quad . \quad . \quad . \quad . \quad (27)$$

per unit of length.

As for the case $\vartheta = \frac{1}{2}\pi$, it will suffice to mention here that the two principal beams are rectilinearly polarized. For the one, whose vibrations are parallel to the lines of force, the maximum of absorption, which occurs when $n = n_0$, has an intensity determined by

$$h_I = \frac{n_0\mu_0\beta}{cg} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (28)$$

For the other beam, whose vibrations are at right angles to the lines of force, the absorption for $n = n_0$ may be calculated by the formula

$$h_{II} = \frac{n_0\mu_0\beta g}{c(4v^2+g^2)} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (29)$$

§ 6. Let us now pass on to consider the propagation in a direction making an angle ϑ with the lines of force. In doing so we shall, however, exclude cases in which this angle is very near 0 or $\frac{1}{2}\pi$, because for these directions some terms which may in general be omitted, might become of influence [1]).

When both $sin\,\vartheta$ and $cos\,\vartheta$ are large in comparison with the small quantities occurring in our calculations, formula (12) may be replaced by

$$\xi^2 - \xi\,\eta\,sin^2\vartheta - \zeta^2\,cos^2\vartheta = 0, \quad . \quad . \quad . \quad . \quad . \quad . \quad (30)$$

so that

$$\xi = \frac{1}{2}\,\eta\,sin^2\vartheta \pm \sqrt{\frac{1}{4}\,\eta^2\,sin^4\vartheta + \zeta^2\,cos^2\vartheta} \quad . \quad . \quad . \quad (31)$$

At the same time (14) becomes

$$\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}} = -i\frac{\zeta\,cos\,\vartheta}{\xi} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (32)$$

We have, therefore, when the quantities relating to the two principal beams are distinguished by the indices I and II,

$$\left(\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}}\right)_I \left(\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}}\right)_{II} = -\frac{\zeta^2\,cos^2\vartheta}{\xi_I\,\xi_{II}},$$

or, on account of (30),

---

[1]) Notwithstanding this, we shall find that, if we put $\vartheta = 0$ or $\vartheta = \frac{1}{2}\pi$, we can deduce from some of our formulae results that are true for a propagation along the lines of force or at right angles to these lines.

$$\left(\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}}\right)_I \left(\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}}\right)_{II} = 1.$$

This means that one of the characteristic ellipses can be considered as the reflected image of the other with respect to a line bisecting the angle $X'OY$, a rule which also applies to the direction of motion in the two cases.

The imaginary parts of u_1, u_{2+}, u_{2-}, on which the absorption ultimately depends, have their maximum values for $n = n_1, n_{2+}, n_{2-}$, and have diminished to a small part only of the maximum value, when $|n-n_1|$, $|n-n_{2+}|$ or $|n-n_{2-}|$ is equal to a moderate multiple of the coefficient g.

From this we can infer that, when v is sufficiently great in comparison with g, there will be three maxima of absorption at the points of the spectrum determined by (16), and that, if v greatly surpasses g, we have three absorption bands that are completely separated, the body being practically transparent to rays of the interlying wave-lengths. At a point where the imaginary part of one of the quantities u_1, u_{2+}, u_{2-} has its maximum value, both the real and the imaginary parts of the two other quantities may, under these circumstances, be neglected in comparison with that maximum value. For $n = n_{2+}$, for instance, we may put

$$u_1 = 0 \quad , \quad u_{2+} = i\frac{\beta}{g} \quad , \quad u_{2-} = 0,$$

$$\eta = i\frac{\beta}{g} \quad , \quad \zeta = -i\frac{\beta}{g},$$

by which the roots of equation (30) become

$$\xi = -\eta \cos^2\vartheta \text{ and } \xi = \eta.$$

Choosing the first root, we find

$$\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}} = -\frac{i}{\cos\vartheta},$$

$$(\mu) = \mu_0\left\{1 - \frac{1}{2}i\frac{\beta}{g}(1+\cos^2\vartheta)\right\},$$

and if we take the second

$$\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}} = i\cos\vartheta,$$

$$(\mu) = \mu_0.$$

It appears from these results that only the first of the two principal rays is absorbed, and that the axes of its characteristic ellipse are parallel to OY and OX', being to each other in the ratio of 1 to $\cos\vartheta$; this ellipse can be considered as the projection on the wave-

front of a circle whose plane is at right angles to the lines of force. If we imagine a point moving along this circle in the direction in which the equivalent electron moves in the mode of motion $\varkappa_2+$, the projection of this motion on the wave-front indicates the direction in which the characteristic ellipse is described.

All this agrees with the elementary theory of the ZEEMAN-effect and similar remarks apply to the other outer component of the triplet.

§ 7. We shall now enter upon some more details concerning the propagation of rays whose frequency n_0 corresponds to the middle point of the triplet. In order not to exclude cases in which the components of the triplet are not neatly separated or hardly so, we shall *not* assume that v is much greater than g.

For $n = n_0$ we may use the values (24) and (25), whereas

$$\eta = -\frac{2iv}{g}\zeta \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (33)$$

Hence, if we abbreviate by putting

$$\frac{v}{g}\frac{sin^2\,\vartheta}{cos\,\vartheta} = q, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (34)$$

we find from (31) and (32)

$$\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y} = q \pm i\sqrt{1-q^2}\,, \quad . \quad . \quad . \quad . \quad . \quad (35)$$

and

$$\xi = -i\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y}\cdot\frac{4\beta v\,cos\,\vartheta}{4v^2+g^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (36)$$

In discussing these results we shall suppose the quantities v and q, which are related to each other in the manner shown by (34), to be positive [1]).

The nature of the phenomena that will be observed greatly depends on whether q is greater or less than 1. Both cases may occur. Indeed, if we determine an angle ϑ_1 by the equation

$$\frac{v}{g}\frac{sin^2\,\vartheta_1}{cos\,\vartheta_1} = 1, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (37)$$

as we can always do, whatever be the value of $\frac{v}{g}$, we shall have $q > $ or < 1, according as $\vartheta >$ or $< \vartheta_1$.

[1]) The quantity ν is positive when the magnetic field has the direction of the positive axis of z (so that the direction of the rays makes a sharp angle with the lines of force) and when, besides, the right- and left-handed circularly polarized components of the spectral line in the longitudinal ZEEMAN-effect have the ordinary relative positions. The sign of ν is changed both by an inversion of this relative position and by an inversion of the field.

In the first case, i.e. when the angle between the ray and the lines of force is not too small, the ratio (35) is real, namely

$$\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y} = q \mp \sqrt{q^2 - 1} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (38)$$

The vibrations of the two principal beams will therefore be rectilinear, the angles χ_I and χ_{II} which they make with the axis OX', and which we shall reckon positive in the direction from OX' towards OY, being given by

$$sin\ 2\chi_I = sin\ 2\chi_{II} = \frac{1}{q} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (39)$$

Both angles lie between 0 and $\frac{1}{2}\pi$, and the smaller of the two, which we shall call χ_I, corresponds to the under sign in (38), so that we may write

$$\left(\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y}\right)_I = q + \sqrt{q^2 - 1},$$

$$\left(\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y}\right)_{II} = q - \sqrt{q^2 - 1}.$$

Equation (36) shows that ξ has now an imaginary value for both principal beams, and, since the same is true of $u_{2+} + u_{2-}$, we see from (22) and (7) that the two beams have the same real index of refraction μ_0 (and therefore the same velocity of propagation), but different indices of absorption, namely

$$h_I = \frac{n_0 \mu_0 \beta}{c(4v^2 + g^2)} \{g + 2(q + \sqrt{q^2 - 1})\, v\, cos\, \vartheta\}, \quad . \quad . \quad (40)$$

$$h_{II} = \frac{n_0 \mu_0 \beta}{c(4v^2 + g^2)} \{g + 2(q - \sqrt{q^2 - 1})\, v\, cos\, \vartheta\} \quad . \quad . \quad . \quad (41)$$

It appears from this that the absorption is strongest for the beam whose vibrations make the smaller angle with the lines of force. This might have been expected on the ground of the elementary theory of the ZEEMAN-effect.

The difference between the expressions $q + \sqrt{q^2-1}$ and $q - \sqrt{q^2-1}$ which occur in h_I and h_{II} increases as q becomes greater. Now, if for a fixed value of $\frac{v}{g}$, the angle ϑ is made to approach the limit $\frac{1}{2}\pi$, (34) shows that q increases indefinitely. When it has become very great, we may replace $q + \sqrt{q^2-1}$ by $2q$, and since $q\, cos\, \vartheta$ tends towards the value $\frac{v}{g}$, as may be seen from (34), we have at the limit

$$h_I = \frac{n_0\, \mu_0\, \beta}{cg},$$

agreeing with the value (28) which we have given for a ray perpendicular to the lines of force and having its vibrations along these lines. At the same time $q - \sqrt{q^2-1}$ may be replaced by $\frac{1}{2q}$, so that h_{II} approaches the limit (29).

On the other hand, when ϑ is made smaller and ultimately becomes equal to ϑ_1 ($q = 1$), both h_I and h_{II} have the limiting value

$$h = \frac{n_0 \mu_0 \beta}{c} \frac{2v \cos \vartheta_1 + g}{4v^2 + g^2}, \quad \ldots\ldots (42)$$

or, if (37) is taken into account,

$$h = \frac{n_0 \mu_0 \beta}{cg} \frac{2v^2 \sin^2 \vartheta_1 + g^2}{4v^2 + g^2}.$$

This lies between the values (28) and (29).

As for the directions of the vibrations in the principal beams, these are determined by $\chi_I = 0$ and $\chi_I = \frac{1}{4}\pi$ in the extreme cases $\vartheta = \frac{1}{2}\pi$ and $\vartheta = \vartheta_1$. The former of these results was to be expected, and the latter shows that for $\vartheta = \vartheta_1$ both directions coincide with the line bisecting the angle $X'OY$. We shall denote this line by OL.

§ 8. It appears from what precedes that for $\vartheta > \vartheta_1$ the state of things is wholly different from the one existing when $\vartheta = 0$, which is characterized by a circular polarization of the principal beams. The transition between these phenomena is formed by those which are observed when $\vartheta < \vartheta_1$.

In this case $q < 1$, so that we may put

$$q = \frac{v}{g} \frac{\sin^2 \vartheta}{\cos \vartheta} = \cos \omega ,$$

by which some of our formulae are simplified. The mode of vibration of the principal beams is determined by the relation

$$\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y} = e^{\pm i\omega} , \quad \ldots\ldots\ldots (43)$$

following from (35), and we may therefore say that if we have at some point of the system

$$\mathfrak{D}_y = a e^{i(nt+p)},$$

with real a and p, the other component of the dielectric displacement will be given by

$$\mathfrak{D}_{x'} = a e^{i(nt+p\pm\omega)}.$$

Taking the real parts of these expressions, namely

$$\mathfrak{D}_{x'} = a\cos(n\,t + p \pm \omega) \quad , \quad \mathfrak{D}_y = a\cos(n\,t + p),$$

we may conclude that both principal beams are elliptically polarized, with the same characteristic ellipse, one of whose axes has the direction of the line OL mentioned above. The difference between the two beams lies merely in this, that the characteristic ellipses are described in opposite directions. In order to see this, we have only to observe that, if $\varkappa$ is the angle between $\mathfrak{D}$ and OX',

$$tg\,\varkappa = \frac{\cos(nt + p)}{\cos(nt + p \pm \omega)},$$

$$\frac{1}{\cos^2\varkappa}\frac{d\varkappa}{dt} = \pm\frac{n\sin\omega}{\cos^2(nt + p \pm \omega)}.$$

In the beam to which the upper signs refer, the direction of the motion corresponds to that of propagation. For this reason we shall distinguish all quantities relating to it by the index + and those which relate to the other beam by the index —.

We need hardly add that the characteristic ellipse coincides with the straight line OL when $\vartheta = \vartheta_1$, and that it becomes a circle when $\vartheta = 0$.

We can further deduce from (43), (36) and (22)

$$\left.\begin{aligned} \mu_+ &= \mu_0\left(1 + \frac{2\beta v\cos\vartheta}{4v^2+g^2}\sin\omega\right), \\ \mu_- &= \mu_0\left(1 - \frac{2\beta v\cos\vartheta}{4v^2+g^2}\sin\omega\right), \end{aligned}\right\} \quad \ldots\ldots \quad (44)$$

$$h_+ = h_- = \frac{n_0\mu_0\beta}{c(4v^2+g^2)}(g + 2v\cos\vartheta\cos\omega),$$

showing that, for $n = n_0$ and for any direction between $\vartheta = \vartheta_1$ and $\vartheta = 0$, the two principal beams are equally absorbed, just like the two circularly polarized beams in the extreme case $\vartheta = 0$. The common index of absorption, for which we shall henceforth write h, diminishes as ϑ increases; for $\vartheta = \vartheta_1$ ($\omega = 0$) it takes the value (42), and for $\vartheta = 0$ ($\omega = \frac{1}{2}\pi$) the value (26). How far these extreme values are different, depends on the relative magnitude of v and g.

§ 9. The difference between the velocities with which a left-handed and a right-handed circularly polarized beam travel along the lines of force, leads to the well known rotation of the plane of polarization. On account of the unequality of the velocities of propagation determined by (44), there is a similar rotation in the interval from $\vartheta = 0$ to $\vartheta = \vartheta_1$, with some difference in the details, however, owing to

the fact that the principal beams are not circularly, but elliptically polarized.

Let one of the beams be represented by

$$\mathfrak{D}_{x'} = a\, e^{-h\,z' + i\left[n_0\left(t - \frac{\mu_+ z'}{c}\right) + p + \omega\right]},$$

$$\mathfrak{D}_{y} = a\, e^{-h\,z' + i\left[n_0\left(t - \frac{\mu_+ z'}{c}\right) + p\right]},$$

and the other by similar expressions containing μ_- instead of μ_+ and $-\omega$ instead of $+\omega$. Or, in other terms, let us write for one beam

$$\mathfrak{D}_{x'} = a\, e^{-hz'} \cos\left[n_0\left(t - \frac{\mu_+ z'}{c}\right) + p + \omega\right],$$

$$\mathfrak{D}_{y} = a\, e^{-hz'} \cos\left[n_0\left(t - \frac{\mu_+ z'}{c}\right) + p\right],$$

and for the other

$$\mathfrak{D}_{x'} = a\, e^{\ hz'} \cos\left[n_0\left(t - \frac{\mu_- z'}{c}\right) + p - \omega\right],$$

$$\mathfrak{D}_{y} = a\, e^{-hz'} \cos\left[n_0\left(t - \frac{\mu_- z'}{c}\right) + p\right].$$

Then, compounding the two, and putting

$$\psi = \frac{n_0}{2c}(\mu_- - \mu_+),$$

we get

$$\mathfrak{D}_{x'} = 2a\, e^{-hz'} \cos(\psi z' + \omega) \cos\left[n_0\, t - \frac{n_0}{2c}(\mu_+ + \mu_-)\, z' + p\right],$$

$$\mathfrak{D}_{y} = 2a\, e^{-hz'} \cos \psi z' \cos\left[n_0\, t - \frac{n_0}{2c}(\mu_+ + \mu_-)\, z' + p\right].$$

Hence, at any point z', the resultant vibration is rectilinear, and its amplitude, considered as a vector, may be represented by

$$2\, a\, e^{-hz'}\, \mathfrak{A},$$

$\mathfrak{A}$ being a vector in the wave front with the components

$$\mathfrak{A}_{x'} = \cos(\psi\, z' + \omega) \quad , \quad \mathfrak{A}_y = \cos \psi\, z'.$$

If this vector is drawn from a fixed point, its extremity describes an ellipse when z' is made to increase continually. The corresponding rotation of the plane of polarization is similar to the one observed in more familiar cases inasmuch as it goes on in a constant direction, but when z' is made to increase at a constant rate, the velocity of the rotation is variable. Its changes are determined by

the rule that the vector $\mathfrak{A}$ describes equal areas in equal times; consequently, the velocity of rotation is greatest when the vibration has the direction of the minor axis of the ellipse.

Let $\varkappa$ be the angle between the vector $\mathfrak{A}$ and the axis of x'. Then the above formulae give the following value for the rotation of the plane of polarization per unit of length

$$\frac{d\varkappa}{dz'} = \frac{\psi \sin \omega}{1 + \cos \omega \cos (2\psi z' + \omega)}, \quad \ldots \quad (45)$$

an expression that is constant only for $\vartheta = 0$ ($\omega = \frac{1}{2}\pi$). For any other value of the angle ϑ we may also consider the *mean* value of the rotation. As the vector $\mathfrak{A}$ makes a complete revolution while z' increases by $\frac{2\pi}{\psi}$, we find for this mean rotation

$$\psi = \frac{n_0}{2c}(\mu_- - \mu_+) = -\frac{n_0\mu_0}{c} \cdot \frac{2\beta v \cos \vartheta \sin \omega}{4v^2 + g^2}.$$

It takes the value (27) for $\vartheta = 0$ ($\omega = \frac{1}{2}\pi$) and vanishes for $\vartheta = \vartheta_1$ ($\omega = 0$).

It must be noticed that, even in the neighbourhood of this latter direction of propagation, whereas the mean rotation per unit of length becomes very small, the rotation $\frac{d\varkappa}{dz'}$ may very well have an appreciable magnitude, if the direction of vibration be properly chosen. In fact, the maximum value of (45) is

$$\frac{\psi \sin \omega}{1 - \cos \omega} = \psi \cot \tfrac{1}{2}\omega = -\frac{n_0\mu_0}{c} \cdot \frac{4\beta v \cos \vartheta \cos^2 \tfrac{1}{2}\omega}{4v^2 + g^2},$$

and this can be of the same order of magnitude as (27), even for a value of ϑ very near ϑ_1 ($\omega = 0$).

The ellipse described by the extremity of the vector $\mathfrak{A}$ is similar in form and position to the characteristic ellipse of which we have spoken in § 8.

§ 10. Summing up the above results (and always confining ourselves to the particular frequency n_0) we may say that in the interval between $\vartheta = \vartheta_1$ and $\vartheta = \frac{1}{2}\pi$ the phenomena are in the main of the same kind as the true transverse ZEEMAN-effect that is observed at right angles to the lines of force; the principal beams present a rectilinear polarization and differ from each other by their indices of absorption, whereas the velocity of propagation is the same for both of them. For values of ϑ smaller than ϑ_1, on the contrary, the effect is similar to the true longitudinal one. In this interval it

is only by their velocities of propagation, but not by the intensity of the absorption, that the two beams are different.

We can diminish ϑ_1 by making v greater. Hence, the region of the transverse effect expands when the magnetic field is strengthened, and finally, when $\frac{v}{g}$ has become very large, i. e. when the distance between the magnetic components far surpasses their breadth, the longitudinal effect is confined to a very narrow interval.

We may add that any definite direction of propagation may be made to fall within the limits of the transverse effect by properly strengthening the field. At the same time, the phenomena become more and more like those that are observed at right angles to the lines of force. Indeed, when v is made greater, q increases continually, as may be seen from (34). The angle χ_I whose value is given by (39), tends towards the limit 0, so that finally the two principal directions of vibration will be perpendicular to each other, the second of them being also normal to the field. Formula (41) shows that, at the limit, the index of absorption of the second beam becomes 0, and equation (40) may be replaced by

$$h_I = \frac{n_0 \mu_0 \beta}{c(4v^2+g^2)} (g + 4qv \cos \vartheta),$$

or, if we take into account the formula (34) and fact that v becomes very much greater than g, by

$$h_I = \frac{n_0 \mu_0 \beta}{cg} \sin^2 \vartheta$$

(cf. formula (28)). These conclusions may be compared with well known results obtained in the elementary theory of the ZEEMAN-effect in the radiated light, namely that the direction of vibration lies in the plane passing through the ray and a line of force, and that the amplitude is proportional to $\sin \vartheta$.

§ 11. From a theoretical point of view it is interesting to examine somewhat more closely the special case in which the rays have the direction determined by the angle ϑ_1. Our formulae would lead us to infer that in this case the two principal beams are polarized in the same way, so that after all there would be only one kind of vibration that can be propagated. Of course this cannot be true. The difficulty can be overcome by pushing our approximations a step farther than has been done in the preceding calculations, in which we have omitted all quantities that are of an order higher than the first with respect to u_1, u_{2+}, u_{2-}.

If we use for η and ζ the exact formulae (19) and (20) — again confining ourselves to the case $n = n_0$ — we still find an imaginary value for the ratio $\frac{\zeta}{\eta}$. We can therefore define a real angle in the first quadrant by the equation

$$\frac{\sin^2 \vartheta_1}{\cos \vartheta_1} = -2i\frac{\zeta}{\eta}, \quad \ldots \ldots \ldots \quad (46)$$

and it is for the direction of propagation determined by *this* angle, that we shall perform the following calculations.

It follows from (33) that the angle ϑ_1 which we introduce now becomes equal to the angle originally denoted by the same symbol when we take for η and ζ their former values. These are a little different from those which we must now ascribe to these quantities, and therefore the direction of propagation assumed in our present calculation does not exactly coincide with the direction which we considered in the preceding article as the boundary between the regions of the longitudinal and the transverse effect. The deviation of one direction from the other is, however, insignificant; it will even be found to be small in comparison with the new terms that will now become of importance.

We shall again begin with the determination of ξ. For this purpose we have to use equation (12), for which, on account of (46), we may write

$$\xi^2 + 2i\,\xi\zeta \cos \vartheta_1 - \zeta^2 \cos^2 \vartheta_1 = -\xi^2 \eta \cos^2 \vartheta_1 - \xi\zeta^2 \sin^2 \vartheta_1 + \eta\zeta^2.$$

Here, the terms on the left-hand side, the only ones with which we were concerned in our first approximation, form a complete square, and this is the reason why we found two principal beams identical with each other.

In the approximation now required we must retain the terms on the the right-hand side, but it will suffice to substitute in them the values of ξ, η, ζ obtained in our former calculation. Distinguishing these by the index 0, we get the following equation for ξ

$$\xi + i\zeta \cos \vartheta_1 = \pm \sqrt{-\xi_0^2 \eta_0 \cos^2 \vartheta_1 - \xi_0 \zeta_0^2 \sin^2 \vartheta_1 + \eta_0 \zeta^2{}_0}. \quad (47)$$

As to the three quantities ξ_0, η_0, ζ_0, the last of them is given by (25), and we have, in virtue of (46),

$$\eta_0 = -2i\zeta_0 \frac{\cos \vartheta_1}{\sin^2 \vartheta_1},$$

and, on account of (36), since the value of $\frac{\mathfrak{D}_{x'}}{\mathfrak{D}_y}$ was 1,

$$\xi_0 = - i \frac{4\beta v \cos \vartheta_1}{4v^2 + g^2} = - i \zeta_0 \cos \vartheta_1.$$

Substituting these values in (47), we get

$$\xi = - i \zeta \cos \vartheta_1 \pm (1 - i) \zeta_0 \delta, \quad . \quad . \quad . \quad . \quad . \quad (48)$$

where we have put

$$\delta = \frac{1 + \cos^2 \vartheta_1}{\sin \vartheta_1} \sqrt{\tfrac{1}{2} \zeta_0 \cos \vartheta_1}.$$

The new term to which our present approximation leads us, namely $(1-i)\,\zeta_0\delta$, is thus seen to be of the order of magnitude $\zeta_0^{3/2}$, so that it will be allowable to neglect quantities of the order ζ_0^2. Such are: first, the difference between the values of ϑ_1 given by (37) and (46), and secondly, the change that would be brought about in our results if we took into account the difference between η,ζ and η_0,ζ_0. Moreover, we may neglect the products $\xi\zeta$, $\eta\zeta$, and $\xi\eta$ in formula (14), and we may again use for the complex index of refraction the equation (22), substituting in it, on account of (24), (25) and (37),

$$\frac{1}{2}(u_{2+} + u_{2-}) = \frac{i\beta g}{4v^2 + g^2} = \frac{ig}{4v}\zeta_0 = \frac{1}{4} i \zeta_0 \frac{\sin^2 \vartheta_1}{\cos \vartheta_1}.$$

Finally we deduce from (48)

$$\xi = - i \zeta_0 \cos \vartheta_1 \pm (1-i) \zeta_0 \delta,$$

from (14)

$$\frac{\mathfrak{D}_y}{\mathfrak{D}_{x'}} = 1 \mp (1 + i) \frac{\delta}{\cos \vartheta_1},$$

and from (22)

$$(\mu) = \mu_0 \left\{ 1 - \frac{1}{4} i \zeta_0 \frac{1 + \cos^2 \vartheta_1}{\cos \vartheta_1} \pm \frac{1}{2} (1 - i) \zeta_0 \delta \right\}.$$

The result is that in this special case, like in the general one, there are two distinct principal beams, with different characteristic ellipses, both deviating somewhat from the straight line OL mentioned at the end of § 7. Between the two there is a slight difference, both in velocity of propagation and in index of absorption.

The regions of the longitudinal and the transverse ZEEMAN-effect are thus found not to be sharply separated from each other, as we concluded in § 10, but to overlap to a certain extent. This shows that, strictly speaking, the consideration of additional terms of the order $\zeta_0^{3/2}$ is necessary, not only in the case $\vartheta = \vartheta_1$, but also for other directions of propagation lying within a small angle on both sides of the direction determined by the angle ϑ_1.

Botany. — *"On Distylium Stellare O. K. and Aporosa Campanulata J. J. S."* By J. J. SMITH. (Communicated by Prof. F. A. F. C. WENT).

In the "Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling der Koninklijke Akademie van Wetenschappen te Amsterdam van Vrijdag 23 April 1909" (These Proc. May 1909) Dr. S. H. KOORDERS published a note entitled: "Plantae Junghuhnianae ineditae III. Einige pflanzengeographische Bemerkungen über eine im Java'schen Hochgebirge wildwachsende Art von der Hamamelidaceen-Gattung Distylium Sieb. und Zucc."

This paper gives me occasion for the following remarks.

Dr. KOORDERS's note is partly based on the supposition, that I had described in Icones bogorienses III (1907), t. CCXXIX (not t. 209, as Dr. K. cites) a *Mallotus campanulatus*; however I described l. c. an *Aporosa campanulata*.

In the said "Verslag" p. 954 Dr. KOORDERS further writes:

"Im Jahre 1907 wurde dieselbe Art (viz. *Distylium stellare* O. K. von Herrn SMITH unter Benutzung der von BOERLAGE gemachten Bestimmung als *Mallotus* spec. nov.) als neue Art der Euphorbiaceen-gattung *Mallotus*, nämlich als *M. campanulatus* J. J. S. nov. spec. beschrieben und abgebildet. Aus der zitierten Publikation von Herrn J. J. SMITH geht hervor, dass man in Buitenzorg damals (1907) die von OTTO KUNTZE im Jahre 1891 publizierte, sehr gute Speciesdiagnose dieser Art, nämlich *Distylium stellare* O. KUNTZE (Revisio gen. pl. I, 233) übersehen hat, wie auch früher bei der in 1895 ausgegebenen Bearbeitung der *Hamamelidaceae* für die KOORD. en VALETON Bijdrage Boomsoorten Java II, 202, von meinem Collaborateur Herrn Dr. TH. VALETON und mir die erwähnte, in 1891 von O. KUNTZE publizierte Speciesbeschreibung übersehen worden war. Indessen kann ich auf Grund meiner vergleichenden Untersuchung der Leidener, Berliner und Kew-Originalspecimina mit Sicherheit erklären, dass die erwähnte, von Herrn J. J. SMITH als neue Art der *Euphorbiaceae* im Jahre 1907 beschriebene und die von O. KUNTZE in 1891 als neue Art der *Hamamelidaceae* beschriebene javanische Waldbaum-species vollkommen identisch sind."

I regret, that I cannot share Dr. KOORDERS' opinion. When we compare the description and the plate of *Aporosa campanulata* with the characteristics of the genus *Distylium* the result is, that the former cannot possibly belong to this genus. Even the defective description of *Distylium stellare* O. K. suffices to prove, that the two plants in question are totally different, though they may show a

superficial resemblance in the form of their leaves. So the stamens are very differently shaped (in the description of *Aporosa campanulata* Ic. bog. III, 72, a line is missing, but the delineation is very clear), the ovarycells in *Distylium* contain only one, those in *Aporosa* two ovules, and the style in the two genera is distinct. Moreover the inflorescence and leaves (nervation, hairiness, leafstalk etc.) are unlike in both plants.

Distylium stellare O. K. in Herb. Kds. only is represented by ♀ specimens. However I think, that O. KUNTZE placed the plant in the right family and genus.

Aporosa campanulata J. J. S. is a true *Euphorbiacea* and doubtless an *Aporosa*. (The plant probably belongs to the Philippine *A. sphaeridophora* Merr.).

Distylium stellare O. K. in Herb. Kds. is represented from the following places:

"Wildhoutaanplant" near pasanggrahan Pengalengan, 1400 m. above sealevel, afd. Bandoeng, res. Preanger. Therefore probably not of spontaneous growth here.

Forest Bentjana on G. Slamat, 1360—1500 m. above sealevel, afd. and res. Tegal.

Near pasanggrahan Simpar on the northslope of G. Slamat, res. Tegal.

Forest near the summit of G. Prahoe, 2200—2550 m. above sealevel, Dieng-plateau, res. Bagelen. On one of the sheets from this habitat "Dieng-kultuur" is noted.

Forest Mantren, afd. Magelang, res. Kedoe.

"Wildhoutkultuur" on G. Sindoro, Myrica near Kledoeng, 1405 m. above sealevel, res. Kedoe.

The plant was first (1891) designated by Dr. KOORDERS as *Flacourtia* sp. and *Flacourtia* n. sp., the specimen from Pengalengan as? *Aporosa*? Later most of the specimens were labelled *Mallotus* sp. (n. 11), while finally (in 1899) all specimens were named *Aporosa*? *aurita* Baill. The sheet from Mantren bears a label, written in 1898:? *Euphorbiaceae* (?? Genus dubium BOERL. et KDS.).

All these labels are in the handwriting of Dr. KOORDERS.

Aporosa Campanulata J. J. S. is represented in Herb. Kds. from:
Takoka, 1200 m. above sealevel, afd. Tjiandjoer, res. Preanger.
Tjigenting, 1450 m. above sealevel, afd. Bandoeng, res. Preanger.
Forest Grendjeng, near Bringombo, 1800 m. above sealevel, afd. Bandjarnegara, res. Banjoemas.

Dr. KOORDERS determinated the plant rightly as an *Aporosa*,

Botany. — *"The inadmissibility of the statolith theory of geotropism, as proved by experiments of* Miss. C. J. PEKELHARING. By Professor F. A. F. C. WENT.

The statolith theory as formulated by HABERLANDT and NÊMEC in order to explain the perception of gravitational force by plants, has led to a whole series of papers in which the authors have attempted to advance arguments for and against the theory. It is not my intention to discuss these here; it may suffice to say that none of them have led to a definite conclusion concerning the question and have at most induced HABERLANDT (NÊMEC did not occupy himself any further with the matter) to modify the theory several times in order to bring it into line with newly discovered facts.

The adherents of the statolith theory suppose that the perception of gravitational stimuli takes place in the plant cell in consequence of the pressure exerted by starch grains on the protoplasm. We may consider it an entirely open question, whether in this case we should postulate that the starch grains change their place when the orientation of the cell with regard to the perpendicular is changed, but in any case we may demand, that the faculty of perceiving gravitational stimuli should go together with the presence of starch grains. Hence it is intelligible that both HABERLANDT and NÊMEC tried to make the starch disappear from the sensitive parts, the former with the aid of low temperatures, the latter by imbedding the objects in plaster of Paris. They were successful in this; the power of perceiving gravitational force disappeared with the starch and returned when the starch was regenerated. These experiments did not, however, furnish a conclusive proof, because it was uncertain whether the treatment which brought about the disappearance of the starch, did not also affect adversely the reactivity of the protoplasm.

A detailed discussion of these experiments seems superfluous, on account of what follows. In experiments on the perception of gravitational stimuli Miss C. J. PEKELHARING attempted in my laboratory to bring about the disappearance of the starch from the geotropically sensitive parts by a different method from that employed hitherto. In so doing she obtained important results of which I wish to give here a preliminary outline.

An investigation of FLURI [1]) had shown that by means of aluminium salts it is possible to eliminate starch from living cells, both from

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[1]) M. FLURI, Der Einfluss von Aluminiumsalzen auf das Protoplasma, Flora, Bd. 99, p. 81, 1908.

leaf veins and from Algae. This suggested to Miss PEKELHARING that in this way it might also be possible to remove starch from the root cap of living roots. It was at once found, however, while experimenting with the rootlets of *Lepidium sativum*, that aluminium salts are poisonous. The roots generally react with an abnormal growth in thickness and traumatotropic curvatures, by means of which they turn away from the aluminium solution.

Experiments have of late been made with solutions which are so called physiologically balanced, i.e. in which the toxicity of one metallic ion is annulled or at least compensated for by another metal; these experiments led to attempts to counteract the toxicity of the aluminium, an object, which was more or less achieved by using K-ions.

If the roots of *Lepidium* are grown in a solution containing 1 gram of potash-alum in 4 litres of tap-water, it is found that there are a number of roots which become very long and thin and retain their starch, while others thicken in a more or less abnormal manner and lose their starch. Of the latter there are always some which show the above mentioned traumatotropy and which are therefore unsuitable for further experimentation. If these are cut away, a certain number always remain, which are free from starch and straight. If the solution is a little weaker, the traumatotropic rootlets indeed disappear, but the starch then persists; in stronger solutions the curvature is so general, that the material becomes useless.

The presence or absence of the starch can be ascertained by means of chloral-iodine; in the transparent rootlets of *Lepidium* it is then possible to see at once by small magnification, whether or not starch is present; increased certainty is obtained by making longitudinal sections. In order to be absolutely sure, however, a series of microtome sections were made in certain cases, and each section was examined separately for starch; it was thus found that the chloral-iodine test is sufficiently reliable.

The experiment was now carried out in such a way, that the seeds were soaked in water for a day and then placed on gauze which was stretched over a lamp chimney. The chimney-glass was placed in the solution of potash alum in such a way, that the level of the liquid was $^1/_2$ cm. below the gauze. After a few days the rootlets which had grown through the gauze, had attained a suitable length. Those with traumatotropic curvatures were then removed, and the lamp-glass was placed horizontally in order to expose the roots to the stimulus of gravity. All experiments were carried out in the dark, in order to exclude the action of light.

It was now clearly shown, that a number of rootlets, which had curved geotropically, were completely free from starch. Whatever views we may adopt with regard to the harmful action of the aluminium on the plant, or with regard to the fact that by no means all roots were free from starch, the fact, established with certainty, that roots, the tip of which had become starch-free, nevertheless curved geotropically, proves conclusively that the perception of the stimulus of gravity can take place *without* statoliths.

At most the protagonists of the statolith theory may still maintain, that the starch grains could in any case accelerate the perception of gravity. On this point a conclusive answer could only be obtained by determining, the presentation time for geotropism in roots with and without starch. This determination was unsuccessful, for two reasons: In the first place the harmfulness of the solution and the tendency to traumatotropic curvature make it necessary to stimulate somewhat longer in order to get curvatures which can be readily observed and secondly these water cultures cannot be placed on a clinostat during the latent period, a condition which is necessary in the case of this object, in order to obtain definite curvatures with the presentation time.

Utrecht, October 1909.

Physics. — "*The degree of completeness of the circular polarization of magnetically divided lines.*" By Prof. P. ZEEMAN.

1. A luminous gas placed between the poles of an electromagnet observed along the lines of force, gives in the simplest case, two spectral lines of different wavelengths. These lines are situated at both sides of the original line. In accordance with LORENTZ's elementary theory my observations so far published tend to show almost perfect polarisation of the lines of this doublet, the polarization being right-handed for one, left-handed for the other of its components. If the direction of the field is reversed the sign of the polarization becomes opposite.

Corresponding to the doublet, observed parallel to the field, a triplet is seen when the light emitted at right angles to the field is analysed. The components of this triplet are linearly polarized.

LORENTZ as early as 1898 [1]) showed that some conclusions concerning the polarization of the components of magnetically divided

[1]) LORENTZ. These Proceedings June 1898. p. 113. The Theory of Electrons. Teubner. Leipzig. p. 149. 1909.

lines can be drawn from general principles, independently of any particular theory.

One of these results is that light of a definite frequency radiated along the lines of force, can never show a trace of linear or elliptic polarization; it must either be unpolarized, or have a circular polarization, partial or perfect.

LARMOR [1]) in 1900 concluded that perfect circular polarization of the components of the doublet, would prove that the corresponding permanent types of vibration in the molecules are exactly circular.

In order to explain the more complicated magnetic effects VOIGT [2]) specialised the general theory of vibrating electric systems by supposing particular magnetic links between electrons and by introducing the hypothesis that the luminous particles take a definite orientation under the action of the field. A particular direction, the "axis" of each particle, becomes under the action of the field parallel to the lines of force.

A rotation of the particles around this specified axis undoubtedly is possible, at least it is not excluded. This rotation has no influence upon the frequency. The orbits of the electrons however are rotated as also are the orbits of the "equivalent" electrons recently introduced by LORENTZ [3]) in order to simplify the theory of systems containing a number of electrons.

The linear polarization of the light emitted normally to the field, proves that the orbits of the electrons either are straight lines parallel to the lines of force or ellipses in planes perpendicular to the field.

The completeness of the circular polarization parallel to the magnetic force would prove that the ellipses are circles. Partial circular polarization however would prove the existence of ellipses with all possible fortuitous orientations in planes normal to the field.

In general one would expect, that the components of a magnetically subdivided line emit partially polarized light parallel to the field. Without hypothesis or further measurements this more general statement must even be regarded as the most probable. A quantitative examination of the ratio of circularly polarized light contained in the total light emitted by the components, has not been made till now.

2. My observations with ROWLAND's grating are consistent with

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[1]) LARMOR. Aether and Matter. p. 345.

[2]) VOIGT. Magneto- und Elektrooptik, Teubner, Leipzig. p. 98. 1908.

[3]) LORENTZ. Theorie der magneto-optischen Phänomene. Encyclopadie der mathematischen Wissenschaften. V. 3. Heft 2. p. 217. 1909.

the opinion that the radiation of the components of the doublet is accompanied by a rather high percentage of ordinary light.

The brightness of grating spectra is only small compared with the intensity of the original, incident light. The amount of ordinary light contained in the total radiation of the doublet easily could remain below the limiting value necessary for perception, and notwithstanding its intensity compared with that of the spectral line be by no means small.

In order to decide experimentally between the two possibilities, I have made some experiments on the completeness of the circular polarization.

As regards intensity of light MICHELSON's echelon spectroscope certainly surpasses all other spectroscopes of high resolving power. It therefore was the most suitable instrument for the projected experiments and satisfied a first condition I had to attend to.

A second condition to be satisfied for the projected experiments is that the source of light be as intense as possible.

I therefore for the present only investigated the spectra of sodium, mercury and thallium, which can be obtained with great intensity. These few elements moreover have the advantage of exhibiting several different types of magnetic separations.

The methods used for the investigation of the circularly polarized light are given in §§ 3—5. Particulars concerning the sources of light are contained in § 6. The observations and conclusions concerning the chief subject of this paper form the end of this communication §§ 7—13.

3. *Verification of the circular analysers.*

In order to investigate the circularly polarized light of the components it is simplest to use either quarter-wave plates or FRESNEL's rhomb.

The last contrivance has the advantage of suiting simultaneously the necessary conditions for a large part of the spectrum. The quarter-wave plates, however, can be selected only for a very limited part of the spectrum. They can easily be adjusted for the purpose in view rather by splitting mica sheets.

The interference colour exhibited, when the plate is placed between crossed Nicols, in parallel light, gives a measure of the retardation produced.

As the estimation of this colour is, however, always a matter of some difficulty and if a somewhat great accuracy is desired, it is a

much better plan to analyse the light leaving the second Nicol with the aid of a spectroscope. The result becomes more accurate, if one combines a moderately thin plate of double refracting crystal with the mica sheet. The simple theory of the dark bands, now visible in the spectrum, is well known and needs not to be given here. Since the time of FIZEAU and FOUCAULT many physicists have used them for the measurement of phase differences. [1])

A plate of quartz, cut parallel to the optical axis, and of about 2 m.m. thickness, gives 18 bands between the red and violet hydrogen lines.

These bands are most distinct, if the principal sections of the Nicols and the mica be inclined at an angle of 45°.

If the mica and the quartz plate be superposed in such a manner, that the principal sections correspond, a displacement of the bands in a certain sense, e.g. towards the red is observed.

If then the mica be rotated through 90° the displacement is towards the violet.

The amount of the total displacement observed, by the interchange of the two positions of the mica, is easily measured. The ratio of half this displacement and the distance of two succeeding bands gives the difference of phase produced by the mica for the region of the spectrum under consideration.

KÖNIG [2]) and CORNU [3]) suggested the use of a double quarter-wave plate with horizontal boundary line for the easier observation of the longitudinal magnetic effect.

I myself often make use of mica plates of the same kind but divided into three fields, as indicated in the figure. The principal

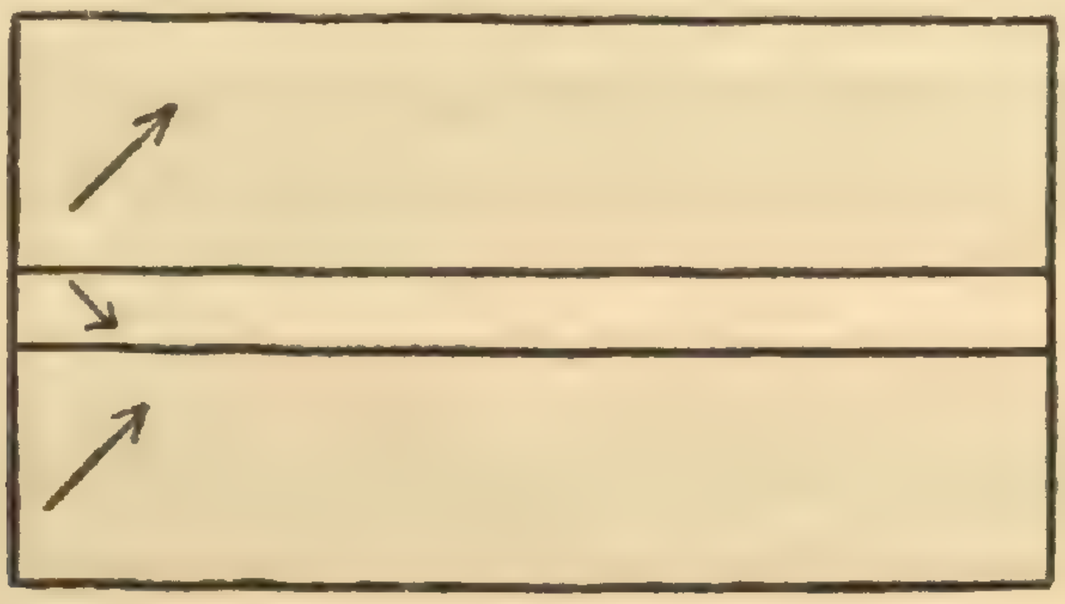

Fig. 1.

sections of the fields are indicated by the arrows.

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[1]) See f. i. CORNU in the paper cited sub 3.

[2]) W. KÖNIG, Wied. Ann. **63** S. 268. 1897.

[3]) A. CORNU. C. R. **125** p. 555. Octobre 1897, Eclairage Electrique **13** p. 246. 1897.

Such a three-fold plate is made nearly as easily by the experimenter as a double one, whereas for measurements its use allows of a somewhat higher accuracy by reason of the symmetry of the field of view.

Using either these double or three-fold mica plates one immediately gets the total displacement of the bands, if a quartz plate be superposed and the combination placed between crossed Nicols. An image of the horizontal lines of separation is projected by means of an *achromatic* lens upon the slit of a spectroscope.

In fig. 1 of the Plate a photograph taken with a three-fold mica-plate is given. The relative retardation produced by the mica is a quarter of a wave-length, if the distance of two bands in the outer fields is just halved by one in the middle field.

A comparison spectrum from $\lambda = 6561$ (H) to $\lambda = 4078$ (Hg) is taken simultaneously. How largely the phase-difference in the mica depends upon the wavelength is very clearly shown.

4. FRESNEL's rhomb gives, as is well known, circularly polarized light after two total reflexions of light polarized at an azimuth of 45°.

This arrangement, if perfect, has the advantage of giving circularly polarized light for all colours of the whole visible spectrum. I examined as to how far the rhomb at my disposition satisfied the requirements imposed by the object I had in view.

The chief purpose of my investigation requires a rather close approach to the ideal condition. For this reason and also because, as far as I know, FRESNEL's beautiful invention was never investigated by the method of interference bands, I may be allowed to give a more extensive treatment of the results than I otherwise would have contemplated.

In order to obtain also with FRESNEL's rhomb two fields in which the bands are shifted in opposite directions, the rhomb is to be combined with a double plate. I used a double-plate [1]) made of a plate of quartz, 1.7 m.m. thick, and cut parallel to the axis. This plate is cut in two halves by a line making angles of ±45° with the principal section; one half is then rotated round an axis perpendicular to the line of separation, through 180°.

Fig. 2 was obtained by means of this double-plate and a rhomb, many years old and belonging to the collection of the Amsterdam laboratory. The deviation from an ideal apparatus is very apparent on inspection of the photograph. Measurement shows that the bands are

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[1]) cf. CORNU l.c.

shifted from the position they should occupy with an accurate quarter-wave apparatus, by an amount, which in the red is nearly 11% of the distance of two succeeding bands. In the green and violet this amount becomes 9 resp. 10.5%.

5. Much smaller deviations gives a recently obtained rhomb, as is readily seen by comparison of the photographs 2 and 3.

A somewhat higher accuracy was still obtained by interchanging the prism of the spectroscope used by a more dispersive one. It now, however, became impossible to photograph the whole spectrum at one operation. Fig. 4 and fig. 5 have been obtained with the old FRESNEL-rhomb and the higher dispersion. With the last dispersion and the new rhomb one can without measurements scarcely decide upon the sense of the error in the relative position of the two band systems (see fig. 6 and fig. 7). The deviation of a band in one field, from the centre of two bands in the other, never exceeds 3.2% of the distance of the bands. (The deviations are 1.7, 2.6 and 3.2% for the red, green and violet).

Let the deviation, defined in this manner, be p percent, then the corresponding error of the phase becomes $\frac{p}{200} \times 360° = 1.8\,.\,p$ degrees. For the green p is about 3%, hence 5°.4. An error of this amount in the phase difference of 90°, which exists between the two linear components into which circularly polarized light can be resolved, may easily be shown to have no influence in the case of the measurements of intensity described in § 7.

Let completely circularly polarized light be incident upon the rhomb, then the light after reflexion by the rhomb, giving a retardation of 90° diminished or increased with a small angle δ, may be represented by:

$$\left.\begin{aligned} x &= a\cos nt \\ y &= a\cos(nt+\delta) \end{aligned}\right\}.$$

Hence

$$x^2 - 2xy\cos\delta + y^2 = a^2\sin^2\delta.$$

The principal axes of this ellipse, become: $\frac{1}{2}\,a\sqrt{2}\sin\delta$ and $a\sqrt{2}$, hence their ratio $\frac{1}{2}\sin\delta$.

When $\delta = 6°$, then $\frac{1}{2}\sin\delta = 0.0522$.

The intensity of the light leaving a Nicol with its plane of vibration perpendicular to the major axis of the ellipse, becomes $(0.0522)^2 = 0.0027$.

As the minimum intensity, which under the circumstances of our observations may be recognized, will appear to be of the order 0.01,

even an error twice or three times the one now accepted for FRESNEL's rhomb, cannot disturb our results.

6. *Sources of light.* Vacuum tubes charged with mercury, thallium or sodium, and heated to the required temperatures, were used as sources of light. The tubes were excited by means of an induction coil with high-speed interruptor.

In order to render the luminosity of the tubes as high as possible, the current in the coil was gradually increased to the maximum just possible without impairing the narrowness of the spectral lines.

In the case of sodium the troublesome separate heating of the tube was eliminated by following a somewhat modified procedure. Mercury tubes require only a preliminary heating, the current once started being sufficient to supply the necessary heat. It was tried to mix up some fine powder of previously heated chloride of sodium with the mercury. The tube now exhibited, besides the mercury lines, the sodium lines with extremely great intensity. Often only some parts of the capillary are radiating; the most suitable part is then selected. In the experiments with sodium the coil was actuated by an alternate current, a maximum of 12 Amp. being possible before the widening of the sodium lines considerably interfered with the work.

7. *Arrangement of the experiments.* A few words will suffice to describe the disposition of the experiments.

A DU BOIS-electromagnet with one perforated polar piece was used. The light emitted parallel to the field and made parallel by means of an achromatic lens, traversed FRESNEL's rhomb, then a second achromatic lens, which forms an image on the slit of the auxiliary spectroscope.

This preliminary analysis enables us to select a definite spectral line for further investigation by means of the echelon spectroscope, a more detailed description of which I gave on a former occasion[1]). Between the second lens and the slit of the auxiliary spectroscope a Nicol's prism was introduced, mounted upon a divided circle, which gave the rotation of the Nicol in degrees.

The front plane of the rhomb was placed accurately perpendicular to the beam.

The plane of incidence of the FRESNEL rhomb was made vertical in most experiments. In a few observations it was set at an azimuth of 45° with the horizontal plane.

In the first case the emergent linear vibrations are at an angle of

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[1]) ZEEMAN. These Proceedings Nov. 30, 1901, p. 247.

45° with a vertical line; in the second case they are either horizontal or vertical.

The method of procedure is very simple. The nearly ideal FRESNEL rhomb transforms light of completely circularly polarized doublets into two perpendicular, linear vibrations. By means of a Nicol either the one or the other of the lines can be completely quenched, *provided there is no trace of ordinary light in the double lines.*

The observations with the lines investigated, indeed show that such must be the case. The value of the result depends upon the sensitivity of the method.

The observations consisted in finding the two positions, on either side of the position of extinction, at which the light was just recognizable with certainty.

If α be the angle the Nicol is then rotated from the zero position, $I \sin^2 \alpha$ represents the brightness of the emergent light, I being the intensity of the linear vibration.

We can be sure that the quantity of ordinary light, emitted by one of the components, must be below $I \sin^2 \alpha$.

8. *Results.* The observations consist in determining the value of α for the different spectral lines. I will arrange the results according to the different types of subdivisions, observed normally to the field.

Triplet, mercury, $\lambda = 5791$, doublet parallel to the field, $\alpha = 7°$ $\sin^2 \alpha = 0.0144$. The observation is somewhat hindered by satellites of the principal lines.

Triplet, $\lambda = 5771$, doublet in direction of field.

In extremely strong fields every component is resolved into three lines. This strength of field was not reached.

$$\alpha = 5° \quad \sin^2 \alpha = 0.0076.$$

Quartet, sodium, $\lambda = 5893$, doublet in direction of field.

$$\alpha = 5° \quad \sin^2 \alpha = 0.0076$$

Sextet, sodium, $\lambda = 5890$, quartet in direction of field.

$$\alpha = 6° \quad \sin^2 \alpha = 0.0108$$

Nonet, mercury, $\lambda = 5461$, strong green mercury line, sextet in direction of field.

$$\alpha = 5° \quad \sin^2 \alpha = 0.0076.$$

Sextet, thallium, $\lambda = 5351$, quartet in direction of field,

$$\alpha = 8° \quad \sin^2 \alpha = 0.0196$$

Probably a lower limit can be obtained, when a more satisfactory thallium tube is made use of.

9. *Influence of reflexions from the walls of the vacuum tube.*

In the course of the investigation different mercury tubes were used. With one of these I had the opportunity of repeating an observation, made on a former occasion [1]), under very less favourable circumstances. I will give some details for a case of simple magnetic separation.

The yellow mercury lines split into two components, if the light is examined parallel to the field. Observing in the latter direction and while using a new tube I was astonished to see the yellow mercury lines divided into *triplets*, the three components being of equal intensity. The plane of incidence of the rhomb was under this observation vertical. The intensity of the central line of the triplet was a maximum when the plane of vibration of the Nicol was horizontal, being zero when it was vertical. The central line apparently is linearly polarized. It is easily seen by means of a figure that this central line may be due to light, emitted nearly at right angles to the direction of the field, and, after being reflected once or twice from the inner surface of the capillary, emerging in the direction of the axis of the electromagnet.

Apparently in the direction of the axis a doublet, circularly polarized, is seen superposed upon a triplet with linear polarization. In the specified position of the FRESNEL rhomb, at an azimuth of 45° of the Nicol one of the components of the doublet, after a rotation of 135° the other component must vanish; then, however, the vertical component of the reflected triplet still must remain visible, its intensity being halved, because the principal section of the Nicol makes in the case now considered an angle of 45° with the vertical line. The outer components of the yellow mercury triplets only have about one half of the intensity of the central component, hence, if expressed in the intensity of the latter only $^1/_4$ remains for the vertical vibrations. Observation wholly confirms this.

If the FRESNEL rhomb be brought into such a position that the plane of incidence makes an angle of 45° with the horizontal plane, the central component must be expected to exhibit circular polarization.

This also was confirmed by observation, as other conclusions to be drawn in the case now under review.

I may be permitted to mention, that the central component in the case of the mercury line 5790 has an asymmetrical position; symmetrical, however, in the case of 5771. This appearance corresponds

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[1]) ZEEMAN, These Proceedings 29 Febr. 1908, p. 573.
LOHMANN, Diss. Halle S. 62, 1907.

exactly to what is observed in a direction normal to the lines of force [1]).

The described phenomena in so far as they are due to the reflected light, ordinarily are only just recognizable, the intensity of the light being rather small. The capillary used in the last mentioned observations was of wide bore. With fine capillaries the intensities of the reflected components are considerably weakened.

The diameter of the luminous filament, which emits the most intense light, and which is visible while the vacuum tube is subjected to magnetic forces, probably has as much influence as the width of the capillary in determining the intensity of the reflexion phenomena under consideration. The results of § 8 have been obtained with tubes exhibiting only slight perturbations due to reflected light.

10. The middle component in one or two of the experiments appeared to exhibit only *nearly* plane polarization. The reason of the very small ellipticity of the reflected light might perhaps be due to the elliptic polarisation, which VOIGT predicted, in the case light is emitted in a direction inclined to the lines of force.

Such obliquely emitted light undoubtedly contributes to intensifying the observed central line.

A somewhat faulty position, however, of FRESNEL's rhomb also may be responsible for a slight ellipticity of the polarization. New experiments of course are wanted before the predicted elliptic polarization can be considered as proved by experiment. In my present observations only a very first trace of the effect may perhaps be operative.

11. Before leaving the subject of réflexions in the tube, one point may be considered somewhat more in detail. Is it possible that the circularly polarized light of the components of the doublet, *reflected* back in the original direction may disturb the purity of the observed effect?

The same electron, which emits right-handed circularly polarized light in the direction of the lines of force, which, let us assume, proceed to us, in the opposite direction sends left-handed circularly polarized light of the same period. By reflexion, however, the sense of the circular polarization is reversed, hence the direct as well as the reflected beams enter the eye right-handed circularly polarized. The

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[1]) ZEEMAN, These Proceedings 30 Nov. 1907. p. 351.

only effect is an increasing of the intensity of the doublet components. It is easily shown that also multiple reflexions from those parts of the wall of the capillary, which are normal to the field, will only modify the intensity.

12. *Experiments with absorption lines.* I have made also some experiments on the sensitiveness of the inverse effect, a sodium flame placed between the perforated poles of a large DU BOIS-electromagnet being traversed by the white light of an arc-lamp. A large concave ROWLAND grating was made use of for the analysis of the light. The ROWLAND grating can be used for the study of the inverse magnetic effect, because the brightness of the arc-lamp more than compensates the feeble reflexion by the grating.

A double quarter-wave plate and a Nicol are traversed by the beam. By means of the mounted Nicol the right-handed circularly polarized component in one part of the field, the left-handed in the other can be made absolutely black. The corresponding position of the Nicol we call the zero-position. In this position not the slightest trace of absorption can be seen at the places of the other components, which only appear after a considerable rotation of the Nicol. The positions of the Nicol, on either side of the zero-position, were found at which the earliest recognition of the absorption lines was possible. The first trace of the lines only became visible after a rotation of 20° from the zero. Hence, we conclude, that the absorption must already be considerable before being perceptible and that ordinary light in the components of the doublet can be recognized more readily by observations on the direct effect.

13. *Conclusions.* I think I have proved by the experiments described 1 that we must accept, till the contrary has been proved, that in the case of *line* spectra the displaced [1]) components of magnetically subdivided lines, emit parallel to the lines of force completely circularly polarized light.

2. that the amount of ordinary light which might be emitted simultaneously with the circularly polarized light, is in the case of several spectral lines less than 1 % of the total intensity of the spectral line.

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[1]) I expressly speak of *displaced* components in order to except a component which has the same position as the unmodified line. The groups of oppositely circularly polarized components then have a central line in common, which must be unpolarized. The quintets $\lambda = 3788.01$ and $\lambda = 3743.45$ of the iron spectrum, investigated by H. BECQUEREL and DESLANDRES and lately measured by Mrs. VAN BILDERBEEK in the Amsterdam laboratory, provide examples of the type. [Note added to the englisch translation].

As the intensity of light of the apparatus used (ROWLAND's grating, then MICHELSON's echelon) has increased, so the percentage of ordinary light, possibly present, has decreased.

We must conclude, that the orbits of the equivalent electrons, in planes normal to the magnetic force, are with great approximation circles. Elliptic orbits, fortuitously distributed in planes normal to the field, need not be conjectured for the representation of the phenomena.

Rather closely connected with the subject of this paper, are the latest investigations of JEAN BECQUEREL [1]) and A. DUFOUR [2]). DUFOUR obtained various new results concerning the banded emission spectra of the alkaline-earth fluorides and chlorides radiating in a magnetic field and in some cases observed incomplete circular polarization. Several absorption bands of xenotime and tysonite according to J. BECQUEREL also exhibited incomplete circular polarization in a longitudinal magnetic field. He showed, however, lately [3]) that in the case of these crystals there is no real incomplete polarization, but that under the action of the magnetic field besides the principal components, others of slightly different wavelength, come into existence, which exhibit a polarization opposite to that of the principal lines.

Physics. — "*On photo- and electrochemical equilibria.*" By Prof. A. SMITS. (Communicated by Prof. J. D. VAN DER WAALS.)

Already previously [4]) I pointed out that if a dark or thermo-chemical equilibrium is exposed to the action of light or electrical energy, and the system is *susceptible* to these forms of energy, a new equilibrium will be established, which deviates from the thermodynamic equilibrium, or in other words which is thermic metastable.

On that occasion the heterogeneous photochemical equilibria in the systems $Ag_2Cl—Cl_2$ and $S—CS_2$, and further the homogeneous photochemical equilibrium $2\,SO_3 \rightleftarrows SO_2 + O_2$ and the homogeneous electrochemical equilibrium $3\,O_2 \rightleftarrows 2\,O_3$ were discussed, in which a photochemical dissociation equilibrium with HCl was predicted.

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[1]) JEAN BECQUEREL C. R. T. **145**, p. 413. 1907.

[2]) A. DUFOUR C. R. T. **146**, p. 118 et p. 229. 1908. Journal de Physique Avril 1909.

[3]) JEAN BECQUEREL. These Proc. p. 146. June 1909. Contribution à la connaissance du phénomène de ZEEMAN dans les cristaux. See also supplement N°. 20. Leyden communications.

[4]) Inaugural Address. Amsterdam Dec. 9th 1907.

Recently this supposition was not only confirmed, but it has appeared to COEHN [1]) that even at the ordinary temperature this equilibrium is so clear and so easy to demonstrate that in future the dissociation of HCl-gas by light can be an experiment regularly made at university lectures.

Moreover of late some other equilibria have been studied in this direction, so that the number of photo- and electrochemical reactions begins greatly to increase.

Owing to the support of a few friends of science, also the Amsterdam laboratory has been able for some time to reconnoitre the new territory of photo- and electrochemical equilibria, and now it can already be stated that the search in this domain has not been in vain.

Before however proceeding to the results of the investigations, which were carried out in conjunction with Dr. ATEN, I will make some preliminary general remarks, from which it will appear, how in my opinion the photo- and electrochemical equilibria may be considered.

For this purpose I shall take the following example. Suppose a high column of an aqueous solution of some electrolyte; if we now lead a hot current of air through this solution, and that in so large a quantity per second that the loss of heat of the current of air during its passage through the solution is very small compared with its total amount of heat, the temperature of the air that leaves the solution will not be appreciably lower than the temperature of the air which enters the solution, and if we now disregard the influence of gravity, we may assume that the liquid column is a homogeneous system with everywhere the same temperature and the same concentration, so that we may apply the law of the active masses to such a system.

If we now expose the column of liquid to dark electrical dischargings or to light, and that in the direction of the longitudinal axis, we do the same thing as just now in so far that we send a current of energy through the column of liquid.

How this current of energy works, need not be discussed here; the result, however, must be that the potentials of the reacting substances are increased, which at constant temperature must exclusively be the consequence of an increase of the internal energy of the molecules.

Not only the external, but also the internal energy of the molecules

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[1]) Ber. **42**, 3183 (1909).

increases with the temperature, and as we can say a priori, that the internal energy of the molecules will no doubt be the principal factor for a chemical reaction, this leads us already to expect, that when electrical energy and light-energy are absorbed, equilibria will be established at the ordinary temperature, which are also obtained with supply of heat-energy, but then at much higher temperature. Now the examples to demonstrate this analogy, are pretty numerous.

REACTIONS.

Supply heat-energy	$3O_2 \rightleftarrows 2O_3$	$N_2 + O_2 \rightleftarrows 2NO$	$2HCl \rightleftarrows H_2 + Cl_2$
„ electrical „	„	„	„ Smits Aten [1])
„ light „	„	„	„ Coehn [2])
Supply heat-energy	$8H_2S \rightleftarrows 8H_2 + S_8$	$Se_x + xH_2 \rightleftarrows xH_2Se$	$2HJ \rightleftarrows H_2 + J_2$
„ electrical „	„ (Smits Aten)	„ (Smits Aten)	„ (Smits Aten)
„ light „	„ „ „	„ „ „	„ „ „
Supply heat-energy	$P_4 + 6H_2 \rightleftarrows 4PH_3$	$As_4 + 6H_2 \rightleftarrows 4ASH_3$	$Sb_x + \frac{3}{2}xH_2 \leftrightarrows xSbH_3$
„ electrical „	„ (Smits Aten)	„ (Smits Aten)	„ (Smits Aten)
„ light „	„ „ „	„ „ „	„ „ „
Supply heat-energy	$2CO_2 \rightleftarrows 2CO + O_2$		
„ electrical „	„ (Smits Aten)		
„ light „	„ (Chapman) [3])		

Thus I might continue to show that really the photo- and electrochemical equilibrium at the ordinary temperature corresponds with the thermic equilibrium of much higher temperature, and in connection with this it is easy to explain that photo- and electrothermic equilibria set in with great rapidity.

This is of importance particularly because we may consider this as a corroboration of our supposition that the most important factor for a chemical reaction is the internal energy of the molecules, which

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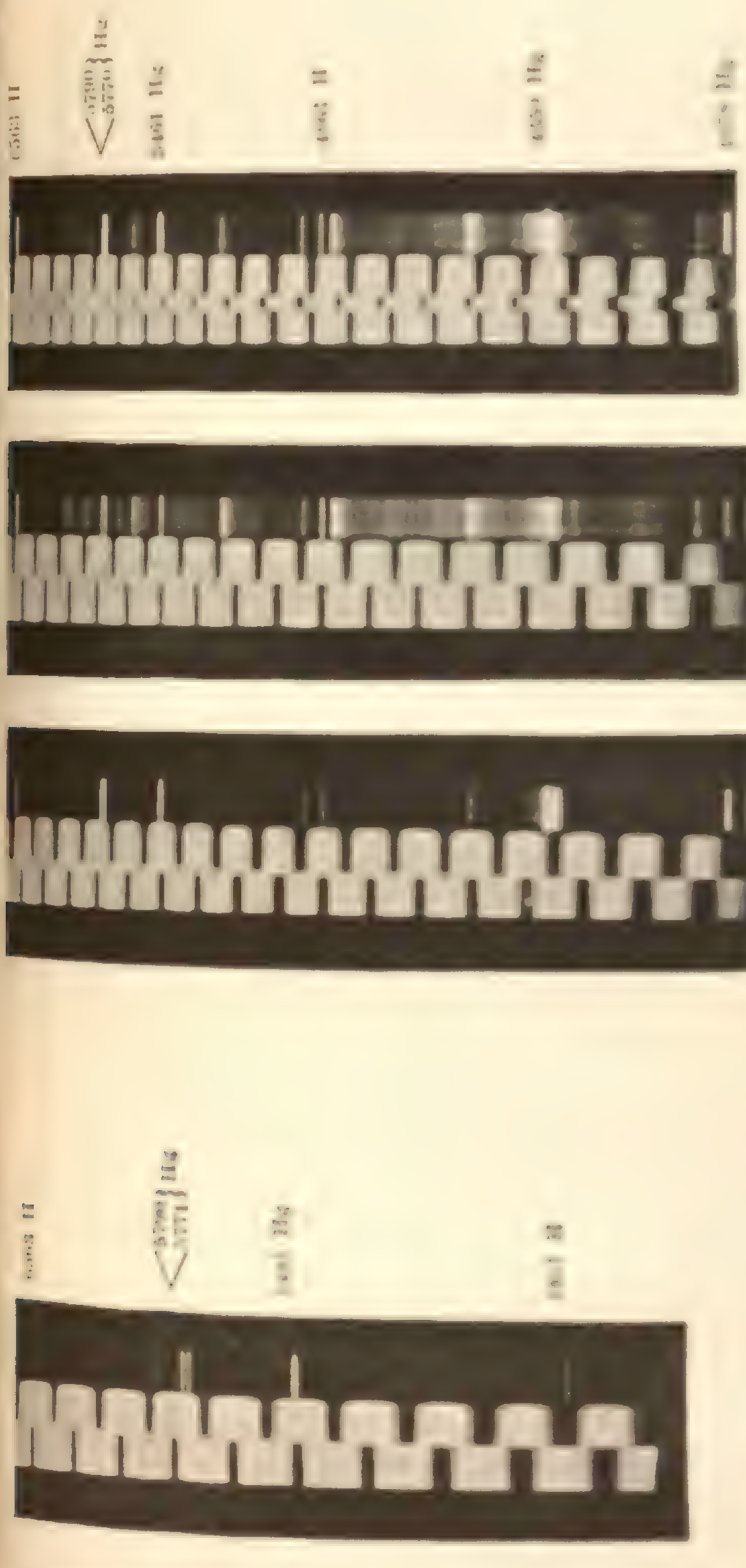
[1]) We will discuss the investigations, some results of which are communicated here, in a following paper.

[2]) l. c.

[3]) Chem. Soc. p. 942 (1907).

P. ZEEMAN. "The degree of completeness of the circular polarisation of magnetically divided lines."



was to be expected, as it is not the molecules, but the atoms or the particles composing the atoms which react chemically.

It is now the question, when may the law of the active masses be applied to a system which is exposed to light energy or electrical energy? The answer to this question is, in my opinion, as follows:

When the quantity of transmitted energy is so great that during the passage of the current of energy no decrease of the energy which is active here, is to be observed, though energy is absorbed by the system, then the system is to be considered as in all respects homogeneous just as in the case mentioned just now of the current of heat-energy, and the law of active masses must be of application.

If, however, the current of energy is less intense, the decrease of energy will be clearly to be demonstrated, the state will differ from layer to layer, and so the system will be non-homogeneous.

So in this case the law of active masses will not be applicable.

It is clear that when the current of energy is too feeble, the equilibrium will be susceptible to a change of the intensity of the current of energy, and so that it is possible to decide in a simple way whether a system at a certain moment may be considered as a homogeneous system or not.

Here we must observe that it must be borne in mind that all kinds of complications may appear; thus it appears among others from DAVIES' investigations [1]) that some component or other may exert influence on the current of energy, which e. g. may give rise to peculiar light phenomena in the case of dark electrical dischargings. It is not surprising that in such a case it seems that the law of active masses does not hold, for the current of energy has been subjected to a change, and what the investigation has revealed is simply this that a change in the properties of the current of energy is of great influence on the situation of the equilibrium.

Since light energy and electrical energy are of a much more complicated nature than heat energy, phenomena may occur in this new domain which were never met with in the thermic region.

In conclusion we may point out that when equilibria are studied at such high temperatures that the vessel in which the system is placed, has become a source of light, we shall get a final state which represents a photo-chemical equilibrium at this high temperature; in a few cases this circumstance will have to be reckoned with.

Amsterdam, October 1909.

Anorg. Chemical Laboratory
of the University.

[1]) Zeitschr. f. physik. Chem. **64**, 657 (1908)

(November 25, 1909).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Saturday November 27, 1909.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van Zaterdag 27 November 1909, Dl. XVIII).

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Anatomy. — *"On the position and displacement of the Foramen magnum in the Primates."* By Prof. L. BOLK.

(Communicated in the meeting of June 26, 1909).

In the Primates the Foramen magnum is very differently situated in the cranial base. Sometimes, as with Mycetes, it lies very near the occipital pole of the skull, so that the condition is found which is the rule with other mammals, sometimes, as in man, it lies about the middle of the cranial base. This latter circumstance has occasionally led to the assumption of a causal relation between the shifting of the Foramen magnum in a facial direction and the acquiring of an erect gait. But HUXLEY already pointed out that the amount of the shifting does not depend on the position which the genus occupies in the Primate system and it will be shown in the present communication that the relation between the two phenomena is certainly not such that the acquisition of an erect gait has been the cause of the displacement of the Foramen magnum.

Together with the variations in position also the inclination of the plane of the Foramen magnum generally changes. This latter point, however, will not be dealt with in this communication, which will only treat of the shifting. The main points of this phenomenon have been known for a long time. In a certain sense the history of comparative craniology was even inaugurated by the study of this displacement. As early as the year 1764, namely, a paper on this subject was published by DAUBENTON [1]), in which he proved the shifting more especially by measuring an angle which has since been known as DAUBENTON's occipital angle, but which has the disadvantage that its two legs do not lie in the median plane. This angle has later been replaced by BROCA's occipital angle which at any rate has the advantage that the two legs are both in the median plane. Yet it seems to me that BROCA's method as well as that which the following investigators (HUXLEY, VIRCHOW, WELCKER) applied in order to express the displacement of the Foramen magnum numerically, may be replaced by a more exact one. They constructed a certain angle which they used as a measure for the displacement. But an analysis of their methods shows that the value of their angle is not only determined by the position of the Foramen magnum but also by different other factors (inclination of the plane of the Foramen magnum.

[1]) Différences de la situation du trou occipital dans l' homme et dans les animaux. Mém. Acad. d. Sciences.

degree of bending of the cranial base, shifting of the anatomical points towards which the legs of the angle diverge). Hence in this communication a different method will be followed which will be presently explained [1]).

This paper contains the first results of an investigation on the anatomy of the Primate skull, especially based on the study of the median plane. In order to prevent repetitions in subsequent communications I shall begin with an enumeration of the materials used. In following papers reference may then be made to this enumeration. All skulls were cut in two along the median plane which was then drawn in natural size by means of the MARTIN pantograph, except the small skulls of Hapale, which were drawn at twice their natural size. The mediagrams of the foetal human skulls were made differently. All measures, ratio numbers, etc. which will be given now and later, have been derived from the mediagrams, unless the contrary is expressly stated.

Human skulls.

20	foetal skulls					
10	skulls from the first year					
5	,,	,,	,,	1st — 2nd	year	
7	,,	,,	,,	2nd — 3rd	,,	
4	,,	,,	,,	3rd — 4th	,,	
5	,,	,,	,,	4th — 5th	,,	
7	,,	,,	,,	5th — 7th	,,	
6	,,	,,	,,	7th — 8th	,,	
4	,,	,,	,,	8th — 10th	,,	
6	,,	,,	,,	10th — 12th	,,	
10	dolichocephalic skulls	(Frisians)		Ind. ceph.	average	75.7
10	,,	,,	(Papouas)	,,	,,	67.5
10	,,	,,	(Negroes)	,,	,,	71.2
10	brachycephalic skulls	(Zeelandians)		,,	,,	84.8
10	,,	,,	(Javanese)	,,	,,	82.7

Besides median sections were made through 10 other dolichocephalic skulls with an average index of 74,8, the origin of which was not accurately known, however.

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[1]) The best work on comparative craniometry of the Primates (that of AEBY) differs from other works also in this respect, that in it ratio numbers are used as much as possible and angular values as little as possible. C. AEBY. Die Schädelformen der Menschen und der Affen. Leipzig 1867.

Skulls of monkeys.

Hapale sp.	6
Cebus fatuellus ♂	5
Cebus fatuellus ♀	5
Chrysothrix sciurea.	10
Mycetes seniculus	5
Pithecia nocturna	4
Ateles sp	7
Inuus nemestrinus	5
Cynocephalus sp	3
Macacus cynomolgus ♂	5
Macacus cynomolgus ♀	5
Cercopithecus sp	4
Colobus guereza.	2
Semnopithecus maurus	5
Siamanga (ad.)	10
Siamanga (juv.).	2
Hylobates sp.	5
Simia satyrus (ad.)	10
Simia satyrus (juv.)	3
Troglodytes niger (ad.)	2
Troglodytes niger (juv.)	2
Troglodytes gorilla (ad.)	8
Troglodytes gorilla (juv.). . . .	2

For Ateles, Cynocephalus and Cercopithecus the data were derived from individuals of different species. The species of the two former genera are not accurately known to me, of Cercopithecus the skulls belonged to the species: albogularis, patas, talapoin and Campbelli. The small skulls of Hapale belonged to individuals from the environs of Paramaribo; it would be difficult to assert whether they all belong to one species. Also of Hylobates I did not know the exact species: two of them originated from the south-western part of Borneo (probably H. concolor), the three remaining ones from Deli on Sumatra (probably H. agilis). But it must be borne in mind that SCHLEGEL, THOMAS, and MAX WEBER are of opinion that these are no distinct species, but regional varities.

The skulls, mentioned here as Frisians, are old "mound" skulls from the first centuries of our aera and hence may be looked upon as representatives of the Germanic skull which may be indicated as skulls of the Homo europaeus, frisius. The above mentioned skulls of Zeelandians were obtained from the drowned land of S. Beveland.

I have shown elsewhere and it had been remarked already formerly by others that in the old inhabitants of S. Beveland we have an almost unmixed round-headed population which must be classed among the short, brown-eyed, brachycephalic race of Europe, now generally known as Homo alpinus. So the skulls may be indicated as skulls of the Homo alpinus zeelandicus [1]).

Before dealing with the Foramen magnum itself I will briefly explain the method followed in this comparative craniological investigation. At the outset the necessity was at once felt of having a base line in thé median plane of the skull, to which the different ratios to be calculated and compared, could be referred. This base line must satisfy certain conditions. Firstly it must join in all skulls two easily determined corresponding points. These points must be determined by the shape of the skull itself. Hence they may not be relief points, since a shifting of these would modify the direction of the base line, while moreover its length would become dependent on the more or less vigorous development of the relief. For the same reason I could not choose a line, the length or course of which could depend on the local thickness of the cranial wall, so that the two terminal points must lie on the interior surface of that wall. Nor did it seem advisable, where skulls of such different form as those of the Primates were concerned, to choose as fixed points the crossing point of one of the sutures intersecting the median plane. For the position of the sutures on the cranial surface depends too much on the development of the adjacent cranial bones themselves. The point where a suture intersects the median plane may be a homologous point with all skulls, but the base line of a comparative craniometric system must lie, if possible, between two homotopic points. [2])

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[1]) By giving this definition I comply with the wish, formulated by Frizzi: Es ist zu wünschen dass alle Länder und deren einzelne Bezirke, welche auf den Alpinus Anspruch erheben werden dürfen, fernerhin dem Oberbegriff Homo alpinus als Unterbegriff den seines näheren Bestimmungsortes beifügen mögen. E. Frizzi. Ein Beitrag zur Anthropologie des "Homo alpinus tirolensis". Mith. Anthrop. Gesellsch. Wien Bnd XXXIX.

[2]) For this reason Klaatsch wrongly asserts that the glabella-lambdaline, proposed by him, is to be preferred to the "glabella-inion" line of Schwalbe. Although the position of the inion may be variable, since it lies in a muscular frame, the lambda is no less variable, being dependent on the surface growth of the squama occipitis. Now when we remember how very variable is the part, played by the ossa interparietalia in the formation of the squama, this would seem a reason why the course of the lambda suture might be even more fortuitous than of any other cranial suture. That moreover Klaatsch's line has no advantage over Schwalbe's became evident to me from what follows. Klaatsch asserts that his Glabella-lambda line in man always forms about a right angle with the Basion-Bregma

The base line of my system, to which all further ratios will be referred, and with respect to which the values of angles will be determined, is indicated in Fig. 1 in the mediagram of a Cebus skull. As frontal point I chose the lowest point of the frontal wall of the skull where the interior surface of the skull bends inwardly in a more or less sharp curve to be continued in the roof of the nasal cavity. The determination of this point presents no difficulties, as a rule, since in the median plane the interior surface of all Primate skulls possesses a distinct frontal wall which may be more or less inclined and may pass more or less gradually into the cranial roof, but which is still always present. I might express this fact as follows: in the median section all Primate skulls possess internally a front. On both sides of the median plane this frontal wall disappears, since the roof of the orbitae approaches very closely under the cranial roof, so that the cranial plane and the orbital roof join under an often very acute angle. It will be shown in a following communication that the human front has its origin not exclusively in a higher vaulting of the frontal bone but also to no small extent in a downward displacement of the eye-sockets. Also in AEBY's work I find this idea already expressed and it seems to me that the investigations of recent years on this part of the skull would have taken a different course if more attention had been paid to the facts in AEBY's work.

The transition of the cerebral into the nasal plane, i. e. in general the foremost lowest point of the anterior surface of the skull, can in most cases be determined at once. Only in a few cases I met with some difficulty in this respect, namely with the skulls of Javanese, since here I found, except in a single case, a frontal crist projecting very far into the cranial space. For the sake of brevity I shall henceforth call this anterior point of the base line the "Fronton". The second point I determined by means of compasses from the first. I namely sought on the posterior wall the point which is at

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line. Now I have measured in the mediagrams of my adult human skulls the angles which the Basion-Bregma line forms with the line of KLAATSCH and with that of SCHWALBE and found the following results. In 70 human skulls the angle which the Basion-Bregma line forms with the glabella-lambda line (KLAATSCH), varies between 84 and 98 degrees and the angle which this same line forms with the glabella-inion line (SCHWALBE) between 104 and 117 degrees. According to the method of KLAATSCH the top of the variation curve lies at 90° (in 17 out of 70 cases), according to the method of SCHWALBE at 112° (in 14 out of 70 cases). From this comparison it is obvious that the proper displacement of the inion is no larger than that of the lambda and that consequently on this account neither of the two lines is to be preferred to the other.

the greatest distance from the fronton. With the lower Primates this point can as a rule be determined at once, in man, however, and especially with juvenile skulls, this is not the case. For it was not unfrequently found that a fairly large part of the interior surface of the skull in the median plane describes a circular arc with the fronton at the centre. Where this was the case the middle of this circular arc was always chosen as the posterior point. In what follows this point will be referred to as the "Occipiton".

The base line extending between fronton and occipiton may at the same time be the line of maximum length within the cranial cavity and is so in the majority of cases, although not always, especially with juvenile skulls. For in man as well as with the Anthropoids the frontal wall of skulls where the milk-teeth are still present, shows a more or less distinctly developed concavity. With such skulls the line of maximum length consequently lies in a slanting direction through the cranial cavity starting about the middle of the frontal bone and ending about the middle of the squama of the occipitale.

Now I shall first point out how in the different skulls I studied the Foramen was placed with respect to the base line, after which I shall compare its position in juvenile and adult skulls of the same species.

It was remarked above that this cannot be done by constructing a certain angle and determining its value and variations. In general it seems to me that in comparative craniology any phenomenon must be studied as little as possible by variations of angular values and that the construction of angles has to be restricted. For any angle requires three points, the two terminals of the legs and the apex. Now a variation in the value of an angle can only then be a true criterion of the course of any phenomenon, when one is convinced that two of these three points have not changed their relative position. And this is hardly to be expected in most cases. The position of every point in the skull varies on its own account, since on every point a large number of factors have a localising influence. Now two points may have a number of these factors in common, but besides several others which are different. Hence if in two skulls the angle between three points is found to vary, we are not justified to explain the difference by the shifting of one of these points only. The inaccuracy inherent in such a method is not entirely avoided, but greatly diminished by constructing projections on a base line instead of making angular determinations and by expressing the course of a phenomenon by the different values of ratio numbers. The degree of accuracy depends with this method

chiefly on the question whether the points, joined by the base line, are really homotopic.

In the present paper this method, which was also followed by AEBY, although his base line was in my opinion wrongly chosen [1]), will be first applied for determining the position of the Foramen magnum. This can be done in a very simple manner. If namely from the anterior edge of the foramen magnum — the basion — a perpendicular is raised on the base line, this latter is divided into an anterior and a posterior part (See fig. 1). The anterior part will in future be indicated by *A*, the posterior by *B*. In my gradually to be developed craniometrical system these letters will always have the same meaning.

The base line itself will be indicated by *G*, so that *A* is the part of *G*, situated before the projection point of the basion, while *B* is the part of *G* behind it. Now *A* becomes greater the further the basion and hence the foramen magnum lies backward. If now we

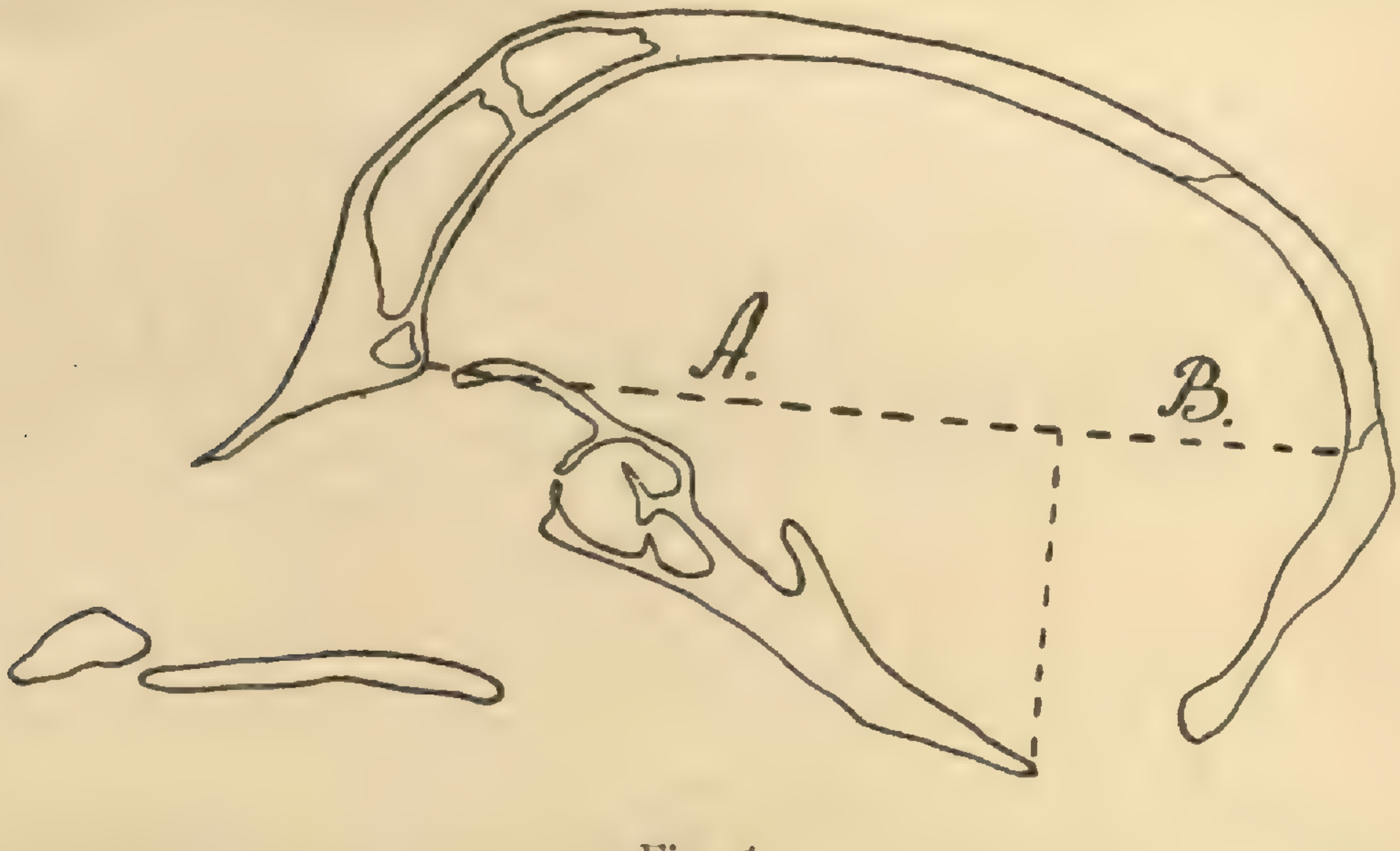

Fig. 1.

put *G* 100, in order to become independent from absolute measures and to be able to make direct comparisons, then by determining *A* and *G* in absolute measure in the mediagram, the ratio of *A* and *G* may be directly expressed by a number which will be larger as *A* itself becomes larger. This ratio number will in future be called the "basal index" of the skull. It is found by the formula

$$\frac{100\,A}{G} = \text{Index basalis.}$$

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[1]) The chief error in AEBY's base line is that he placed it between two "anatomical" points; he derived it from the "structure" of the skull, whereas the base line I have chosen, is determined by its "form".

This index becomes larger as the foramen magnum lies more occipitally.

I shall now first give the basal indices found with the skulls of the different apes, those of man will be given later.

Index basalis of skulls of monkeys.

Hapale 77, 72, 71, 70, 70, 68. Average **71**.
Chrysothrix 62, 61, 60, 60, 60, 59, 58, 58, 57, 56. Average **59**
Cebus 71, 70, 70, 69, 68, 68, 67, 65, 64, 64. Average **67**.
Ateles 67, 66, 65, 64, 61, 58. Average **64**.
Pithecia 76, 75, 74, 70. Average **74**.
Mycetes 95, 91, 86, 84, 78. Average **86**.
Inuus 67, 65, 65, 64, 63. Average **65**.
Cynocephalus 69, 66, 60. Average **65**.
Macacus 69, 69, 66, 65, 65, 63, 62, 60, 60, 60. Average **64**
Cercopithecus 60, 60, 57, 54. Average **57**.
Semnopithecus 76, 76, 74, 71, 70. Average **74**.
Colobus 76, 74. Average **75**.
Siamanga (adult) 78, 77, 76, 76, 76, 75, 75, 75, 73. Average **76**.
Siamanga (juv.) 70, 62.
Hylobates 66, 67, 71, 72, 80.
Chimpanse (adult) 65, 63. Average **64**.
Chimpanse (juv.) 53, 50.
Gorilla (adult) 67, 66, 65, 62, 59, 58, 58, 56. Average **61**.
Gorilla (juv.) 54, 50.
Orang (adult) 69, 65, 60, 60, 58, 58. Average **61**.
Orang (juv.) 55, 53, 51, 51, 50. Average **52**.

Leaving aside for the present the infantile skulls, the basal index appears strongly to vary with the different simian genera, while moreover no inconsiderable individual variations exist. The highest average was found with Mycetes, the lowest with Cercopithecus. At the same time the correctness appears of what had already been asserted by HUXLEY, that the position of the foramen magnum in the cranial base, does not depend on the place occupied by the genus in the system. This is shown most clearly, if the genera are arranged according to the average value, found for their basal index, beginning with the genus with the highest index, where the foramen magnum lies hindmost. The following series is obtained: Mycetes, Siamanga, Colobus, Pithecia, Semnopithecus, Hapale, (Hylobates), Cebus, Inuus, Cynocephalus, Macacus, Ateles, Chimpanse, Gorilla, Orang, Chrysothrix, Cercopithecus.

From this series may also be inferred that there is no direct causal relation between the position of the foramen magnum and the development of the biped gait. Although it will presently appear that the basal index in man is lower than in the other Primates, the given enumeration shows that we must be cautious in attributing this only to the acquisition of an erect gait. If this were the case, Siamanga would certainly not be at the top with the highest basal index but one.

And as to the genus with the highest index value — Mycetes — it is not unlikely that here we have a secondary occipital shifting of the Foramen magnum as the result of the extraordinary development of the hyoid apparatus. The place, occupied by Mycetes, is an exceptional one on account of peculiar developmental phenomena. Taking this into account, Siamanga, the animal which is perhaps more skilled in biped walking than any other Primate, mentioned in this paper, stands at the top as to the value of its basal index and approaches most closely the half-apes of which e.g. Lemur albifrons had an index 87 and Propithecus diadema 80. But one must be careful not to place Siamanga on this ground only lower in the system than the Primates with a smaller basal index and especially not to look upon this phenomenon itself in Siamanga as the preservation of an original condition. Of this I am not convinced. For, as will be shown in a following paper, the position of the foramen magnum is largely determined by the position of the facial skull with respect to the cerebral skull.' And in the Primates an evolutional line in this respect may certainly be recognised, being in short that the facial skull, first situated before the cerebral skull, is displaced under it, after which it is shortened. Now this occipital displacement of the facial skull has a great influence on the position of the foramen magnum.

At the bottom of the series stands with the lowest index Chrysothrix as the representative of the platyrhine monkeys and Cercopithecus of the catarrhine. For these latter an examination of more individuals, specially of the same species, seems desirable to me, in order to settle whether indeed in the genus Cercopithecus the foramen magnum is in general placed so much to the front. The Anthropoids do not, as far as their basal index is concerned, occupy a conspicuously favoured position among the apes and certainly do not form a direct link between the lower apes and man.

The basal index of man is given in the following list, in which the different groups, mentioned above, are kept separate. For the sake of completeness I also mention the cephalic index.

Skulls of Zeelandians (Homo alpinus var. Zeelandicus) Index cephalicus: lowest 81.6, highest 88, average 84.8.

Index basalis: 50, 49, 48, 48, 48, 48, 45, 44, 43, 43, aver. **45.7**.

Skulls of Javanese. Index cephalicus: lowest 78.3 highest 87.9.

Index basalis: 52, 51, 50, 50, 49, 48, 46, 45, 42, 42, aver. **47.9**.

Skulls of Papuas. Index cephalicus: lowest 63.4, highest 69.6 average 67.5.

Index basalis; 46, 45, 45, 45, 44, 44, 44, 44, 43, 42, aver. **44.6**.

Skulls of Frisians (Homo europaeus var. Frisius). Index cephalicus: lowest 71.5 highest 79.1 average 75.7.

Index basalis 48, 47, 46, 46, 46, 44, 44, 44, 42, 40, average **44.7**.

Skulls of Negroes. Index cephalicus: lowest 68.2 highest 76.4, average 71.2.

Index basalis: 50, 49, 48, 47, 47, 47, 45, 44, 43, 42, average **46.6**.

If we compare the index values of man with those of the adult apes, there appears to exist a considerable difference between them, the average of all the human skulls being fifteen units smaller than that of all Anthropoid skulls. By this considerable forward displacement of the foramen magnum the human skull has obtained a characteristic which it shares with none of the other Primates. For if the basal index is greater than 50, this means that the projection of the basion on G. lies behind the middle of this line. Now this was the case without exception with all the adult ape skulls; in man, however, the projection point of the basion lies nearly always before the middle of G and only in exceptional cases it coincides with or lies a little behind it. The individual place occupied by man as to the position of the foramen magnum appears also from the fact that the lowest value I found for the basal index with an adult ape skull, namely 54 with Cercopithecus patas, is higher than the highest value which I found in man: 52 in a Javanese. Whereas with apes A is always larger than B, the opposite is the case with man.

The question whether in man the foramen magnum in the brachycephalic and dolichocephalic skull occupies different positions, corresponding to the difference in shape, must be answered negatively on account of the above figures. To be sure, the two brachycephalic groups, — the Zeelandians and the Javanese — have a slightly higher basal index than the dolichocephalic Papuas and Frisians, but on the other hand the group of the strongly dolichocephalic negroes with their average cephalic index of 71,2 has a basal index which

is higher than that of the very short-headed Zeelandians. [1]) So my results do not indicate that in man there exists a relation between the position of the foramen magnum and the shape of the skull, neither on account of the averages, nor of the range of variation, for with the brachycephalic skulls the basal index varied from 52 to 42 and with the dolichocephalic ones from 50 tot 40.

The independence of cephalic and basal index also appears from the following table in which the two indices are placed side by side for ten long-headed skulls. These skulls do not belong to any of the above-mentioned groups.

Index cephalicus	Index basalis
68.9	46.7
71.5	43.8
72.3	48.—
72.5	41.7
72.9	44.5
73.1	48.7
74.7	50.—
74.8	50.6
75.9	45.5
76.4	44.6

Also in these ten skulls we have again the same range of variation of about 10 units in the basal index as with the five groups above.

The main conclusion from what precedes is that in man the projection of the basion on G regularly falls a little before the middle of this line. Now if we bear in mind that the line of support of the skull, which joins the middle of the two condylae, lies regularly a little behind the basion, the conclusion is arrived at that the projection point of the line of support of the human skull on the base line I assumed, coincides with the middle of this line. It may be taken for granted that in regard to the erect gait of man this relation is a favourable one from a statical point of view.

An unexpected result was obtained by comparing the basal indices of juveniles and adult skulls of the same species. In all cases in which it was possible to make this comparison the basal index of the infantile and juvenile skull proved to be lower than of the adult, in other

[1]) In Aeby's monograph I find the following remark as to the Foramen magnum in negroes: Einige Beobachter glauben die Wahrnemung gemacht zu haben dass die Stellung dieser Oeffnung insofern veränderlich sei, als sie bei gewissen Völkerschaften und zwar speziell den Negern, weiter hinten liege, als bei andern. (Die Schädelformen der Menschen und der Affen. Leipzig 1867 p. 16).

words: *during the postfoetal development of the skull the foramen magnum is shifted occipitally*. I had not expected this result. A priori I should have expected the opposite, namely that the individual development would reflect the phylogenetic. I shall first show the stated for the ape skulls, and then for human skulls.

The direct proof of this shifting is obtained by comparing the basal indices. But also in another manner I will give a striking proof, namely by superposing the mediagrams of youthful and adult skulls.

Of the Siamanga I made sections of two juvenile skulls, the youngest of which possessed all the milk teeth, while no permanent tooth had yet appeared. The basal index of this skull was 62, i. e. eleven units less than the lowest basal index of an adult siamanga skull. The second possessed a mixed set of teeth, the permanent canines, the first premolars and the third molar had not yet come through. Of this skull the basal index was 70. Hence a considerable backward shifting of the Foramen magnum had taken place already, which evidently had not been completed yet.

The difference of position of the Foramen magnum which is indicated by these different values of the basal index, is very clearly shown in fig. 2.

Fig. 2. Mediagrams of an adult and an infantile Siamanga skull.

In this figure the full line is the mediagram through an adult Siamanga skull with a basal index of 76, the dotted one is the mediagram of the young Siamanga with the milk teeth and a basal index of 62. The absolute length of *G* was in the adult skull 80 mm., i. the young one 72 mm. The mediagram of the juvenile skull has accordingly been enlarged in the ratio 80 to 72. If now the base lines are made to coincide, so that the fronton and occipiton of the two skulls are the same, a figure is obtained from which it may be seen at a glance what deformations the skull undergoes when developing from the juvenile to the adult form. In my communications

on this subject I shall repeatedly use such figures; here the method is applied in order to show the difference in position of the Foramen magnum in juvenile and adult skulls.

In fig. 2 this difference is clearly shown for the skull of Siamanga: in the adult skull the Foramen magnum lies considerably more backward than in the juvenile one. At the same time this shifting appears to be chiefly caused by lengthening of the basisphenoid and the basi-occipitale, the base-length of the Fossa anterior appears in youthful individuals to differ little from that in adults.

The backward shifting of the Foramen magnum causes the squama of the occipital to lose its vaulting for the greater part and to stand more vertically.

After this I may be brief concerning the Anthropoids. Of Chimpanse I examined two adult and two juvenile skulls, both with the complete milk teeth. The basal index of the two former amounted to 65 and 63, of the two latter to 53 and 50. Also here the the Foramen magnum consequently appears to lie fairly considerably more frontally in the juvenile skulls. This is moreover shown by fig. 3 in which

Fig 3. Mediagrams of an adult and an infantile skull of Chimpanse.

again the mediagram of a juvenile skull has been superposed on that of an adult one [1]). For the knowledge of the development of the skull it is important to point out that here also the shifting is caused by lengthening of the basi-sphenoid and basi-occipital, together with an increased steepness of the squama of the occipital.

That also with Orang the Foramen magnum is displaced occipitally is sufficiently proved by comparing the two series of basal indices

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[1]) I draw attention to the curious section of the palate of the adult chimpanse which is thickened and inflated by the right and left sinus maxillaris coalescing through the palate, a phenomenon which I also observed in other monkeys (Mycetes).

for adult and juvenile skulls. The proof is here more stringent because of the larger number of juvenile skulls of which the basal index was determined. For the adult skulls I obtained the following figures: 69, 65, 60, 60, 58, 58 and for the juvenile ones 55, 53, 51, 51, 50.

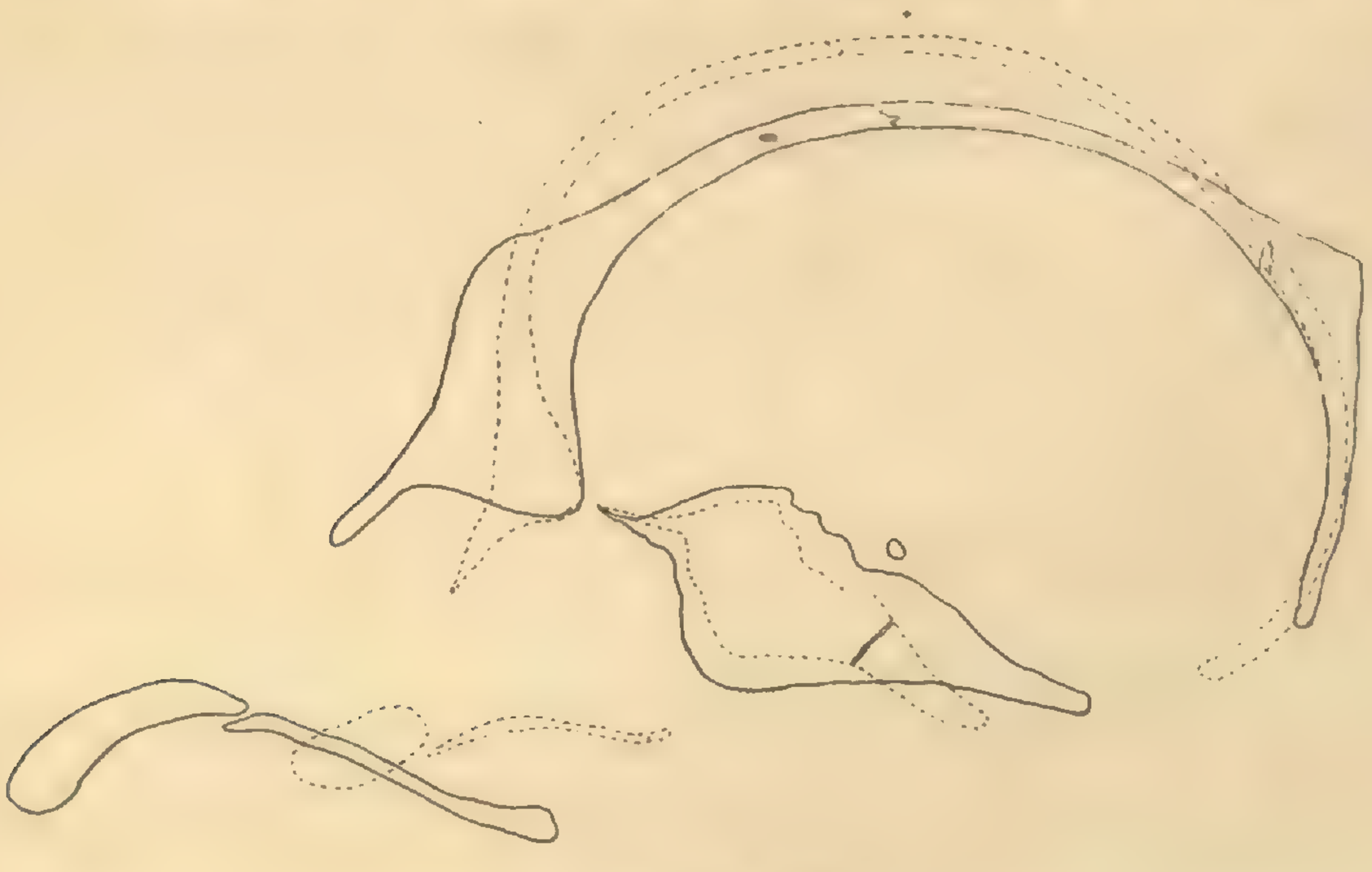

Fig. 4.
Mediagrams of an adult and a juvenile infantile skull of Orang.

In fig. 4 the mediagrams of a juvenile and an adult skull have again been superposed. The former had still all its milk teeth.

For Gorilla finally I refer fo Fig. 5 in which the mediagram of an infantile gorilla skull with complete milk teeth has been drawn on that of an adult skull. We state the same phenomena as with the

Fig. 5.
Mediagrams of an adult and an infantile skull of Gorilla.

remaining Anthropoids: lengthening of the cranial base and diminished vaulting of the occipital are here also met with. As to the basal

index it was found, that in the adult skulls it varied between 67 and 56, while the index value of the juvenile skulls amounted to 50 and 54. With the latter of these two the first molar had already appeared, while the medial incisor had been changed.

By what precedes it is sufficiently proved that in all Anthropoids the foramen magnum during the juvenile period is shifted in a backward direction. At what moment this shifting begins and whether it does so immediately after birth I cannot decide, having no skulls at my disposal in which the milk teeth are not already fully developed.

Several investigators have of late years pointed out that the infantile skull of the Anthropoids stands morphologically nearer the human skull than that of the adult Anthropoids. This phenomenon has especially been discussed in regard to the frontal vaulting. As I have the intention systematically to investigate the whole skull this point will also in due time be dealt with; the remark may here suffice that also in regard to the foramen magnum, this greater agreement is confirmed. For since the basal index of the infantile skull is without exception lower than that of the adult one, it comes nearer the index of the human skull. This approach is so close that the lowest index of the infantile skull of the Anthropoids, namely 50 with Chimpanse, Gorilla and Orang, is equal to the highest I found with Zeelandians and negroes, while with Javanese even a higher index was found. Very important and instructive is a superposition of the mediagrams of the young Gorilla skull with basal index 50 and a Zeelandian skull of the same index, which will be given in a following communication.

There remains to be shown that the same phenomenon which was stated for the skull of the anthropoid apes obtains in man and that here also the basal index of the child is lower than that of adults. The foetal skulls will be left out of account here and only the results obtained with infantile skulls will be given. They will be given as averages for the groups mentioned in the enumeration of materials.

Basal index of the skulls of children.

10	skulls	of	0— 1	year:	average	**41.99**
5	,,	,,	1— 2	,,	,,	**40.4**
7	,,	,,	2— 3	,,	,,	**40.4**
4	,,	,,	3— 4	,,	,,	**41.5**
5	,,	,,	4— 5	,,	,,	**40.—**
7	,,	,,	5— 7	,,	,,	**40.7**
6	,,	,,	7— 8	,,	,,	**40.9**
4	,,	,,	8—10	,,	,,	**44.—**
6	,,	,,	10—12	,,	,,	**43.5**

When interpreting these figures we must take into account that the average I found for five groups of adult skulls was: 44.6, 44.7, 45.7, 46.6 and 47.9. If we compare with these figures the values found for the skulls of children, it is seen at once that in man as well as in the anthropoid apes the foramen magnum is shifted occipitally during development. If the number in each group of infantile skulls were more numerous one would sooner feel inclined to draw the conclusion from the averages found that this shifting begins with the eighth year, about simultaneously with the changing of teeth.

Having learnt how to express numerically the position and displacement of the foramen magnum in the Primates, we may now continue our investigation with the determination of the variations of the inclination of the plane of this aperture. This will form the subject of the following communication.

Physiology. — *"On the changes in the blood serum of sharks after bleeding."* By Mr. F. J. J. BUIJTENDIJK. (Communicated by Prof. H. ZWAARDEMAKER).

(Communicated in the meeting of September 25, 1909).

The blood serum of sharks has an osmotic pressure about equal to that of the salt water in which they live and which varies as the concentration of the salts of the sea or aquarium is changed (RODIER [1]) BOTAZZI [2])).

This osmotic pressure, measured by the lowering of the freezing point of the serum, is partly caused by the salts, dissolved in the blood liquid and for the rest by the urea which is present in high concentration in the body liquids of these Selachians. SCHROEDER [3]) found in the blood of Scyllium with large freshly caught animals 2—3 % urea, which figures have been confirmed by other investigators (BAGLIONI) [4]).

In relation with my investigations on the diuresis of sharks I put myself the question whether perhaps the blood serum would after

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[1]) RODIER, Travaux de la Station Zoölog. Arcachon 1899 p. 103.
[2]) BOTAZZI, Ergebnisse de Physiologie 1908.
[3]) v. SCHROEDER, Zeitschr. f. physiol. Chemie 14 p. 5 75, 1890.
[4]) BAGLIONI, Hofmeisters Beiträge 9 p. 50.

repeated bleeding contain considerably less urea, than is normally the case.

From the dorsal artery of the animals, with which I experimented, the blood was collected, from which after defibrination with glass beads the blood serum was obtained by centrifugation. Only then the resistance and the freezing point were determined by the usual methods. Part of the serum was then tested for its percentage of urea. This was done by determining by KJEHLDAHL's method the N-percentage after the albumen had been removed. From an oral communication of Dr. BURIAN I knew that in albumen-free blood serum and in the urine of Scyllium no other N-compounds than urea are present in measurable quantities.

The albumen was precipitated a few times by HOFMEISTER's method, in which sodium acetate and ferric chloride are added to the liquid and then a precipitate is formed by heating with a little sodium carbonate. The filtrate was absolutely clear and free from albumen. The value obtained in this way for the percentage of urea was smaller, however, than when the albumen was precipitated cold by alcohol and acid or according to BAGLIONI by a hydrochloric solution of asaprol.

Many times the ash percentage of the serum was tested by turning all the salts into sulphates by means of nitric and sulphuric acid and weighing. It is clear that a comparison of the lowering of the freezing point and the ash percentage indirectly gives an idea of the urea concentration of the serum.

In the same way the ash percentage and resistance give an idea of the quantity of albumen, since with equal quantities of dissolved salt the electric resistance of the liquid can only change by varying the quantity of non-electrolytes (colloids).

In the following summary of the experiments Δ means the freezing point, always determined in the same way with a bath temperature of $-5°$ to $-6°$, an undercooling of $-0.5°$, while the stirring rod moved up and down about twice per second. The freezing was induced by inoculating with a splinter of ice.

R is the resistance expressed in Ω, determined according to KOHLRAUSCH's method and calculated by means of OBACHTS' tables. U is the percentage of urea (behind the figure is indicated by H or A whether the albumen was precipitated by HOFMEISTER's method or by asaprol), S is the salt concentration expressed as sulphates, A the albumen.

EXPERIMENT I.

Scyllium canicula ? kilogr., in the aquarium since three days.
Blood serum (14 cc.).

$\Delta = -2.296°$ $R = 37.584\ \Omega$ at $25.4°$ $A = 5.5\ ^0/_0$ $S = 2.019\ ^0/_0$

after 24 hours the animal is killed, serum quite clear.

$\Delta = -2.271°$ $R = 37.148\ \Omega$ at $25.3°$ $A = 4.13\ ^0/_0$ $S = 2.18\ ^0/_0$

EXPERIMENT II.

Scyllium canicula; 2 kilogr., having had no food for 4 days.
Blood serum (25 cc.).

$\Delta = -2.336°$ $R = 34.301\ \Omega$ at 25,3°

after 24 hours 25 cc. of blood serum is obtained.

$\Delta = -2.326°$ $R = 33.2268\ \Omega$ at $25.2°$

blood serum obtained after 24 hours.

$= -2.291°$ $R = 27.309\ \Omega$ at $25.3°$.

EXPERIMENT III.

Scyllium canicula; 2.3 kilogr.
Blood serum (30 cc).

$\Delta = -2.316°$ $R = 32.459\ \Omega$ at $25.1°$ $S = 2.27\ ^0/_0$

animal killed after 24 hours, the blood serum is

$\Delta = -2\ 311°$ $R = 31.01\ \Omega$ at $25.1°$ $S = 2.438\ ^0/_0$

EXPERIMENT IV.

Scyllium canicula; 2.5 kilogr.
Blood serum (25 cc.); $U = 2.07\ ^0/_0\ (H)$

$\Delta = -2.326°$ $R = 37.25\ \Omega$ at $25.1°$ $U = 2.32\ ^0/_0\ (A)$

after 24 hours; blood serum (30 cc).

$\Delta = -2.336°$ $R = 35.68\ \Omega$ at $25.1°$ $U = 2.26\ ^0/_0\ (A)$

after 24 hours; blood serum (20 cc).

$\Delta = -2.326°$ $R = 34.0178\ \Omega$ at $25.1°$ $U = 2.146\ ^0/_0\ (A)$

after 24 hours, animal killed; blood serum

$\Delta = -2.301°$ $R = 31.029\ \Omega$ at $25.1°$ $U = 1.967\ ^0/_0\ (A)$
$U = 1.64\ ^0/_0\ (H)$

EXPERIMENT V.

Scyllium canicula; 1.5 kilogr., in the aquarium since two days.
Blood serum (30 cc).

$\Delta = -2.331°$ $R = 35.4035\ \Omega$ at $25.2°$ $S = 2.06\ ^0/_0$ $U = 2.9\ ^0/_0\ (A)$

killed after 24 hours; blood serum

$\Delta = -2.296°$ $R = 30.975\ \Omega$ at $25.1°$ $S = 2.503\ ^0/_0$ $U = 2.33\ ^0/_0\ (A)$

These experiments prove:

a. The osmotic pressure of the blood serum sometimes remains the same after loss of blood, but generally sinks a little.

b. The percentage of albumen is diminished (as is seen from exp. I with the small loss of blood and from the change in resistance in exp. IV and II).

c. The resistance is diminished by bleeding.

d. The ash percentage of the serum increases; since the osmotic pressure remains unchanged or sinks, this means that:

e. the urea percentage of the blood serum decreases after loss of blood, but does not sink below 1.967 % when the albumen had been precipitated by means of asaprol, or below 1.64% when HOFMEISTER's method had been applied.

Moreover I could state that the blood of animals that had suffered a severe loss of blood, (exp. IV and II) coagulated much more quickly than normally. This phenomenon has also been stated with mammals after bleeding [1]).

I think we are justified to conclude that after loss of blood the serum of sharks is diluted by a liquid which is richer in salt and less rich in urea. Perhaps this liquid is salt water from the sea, perhaps lymph which is in its turn supplemented by sea water; but at all events, even after the severest loss of blood a rather high percentage of urea remained in the serum. It seems that this serum is supplied by the liver. In exp. IV after bleeding the liver was extirpated and the urea percentage determined. I found it to be 1.16 %, while SCHROEDER gives 1.36 %.

Physiology. — *"On the constitution of the urine of sharks with normal and increased diuresis."* Bij Mr. F. J. J. BULTENDIJK. (Communicated by Prof. H. ZWAARDEMAKER).

(Communicated in the meeting of September 25, 1909).

The urine of sharks, as is the case with most cold-blooded animals, has a lower osmotic pressure than the blood serum. Also in man the urine is under certain conditions more diluted than the serum: in some cases of human polyuria, namely, the blood was found to be of normal concentration ($\Delta = -0.56°$), the urine on the other hand to be very much diluted ($\Delta = -0.22°$ to $-0.17°$ C.). METZNER [2]

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[1]) VON DEN VELDEN, Arch. f. exper. Path. u. Pharm. 1909.

[2]) METZNER, Nagels Handbuch der Physiol. II. Bd. 1e H.

says, that this polyuria is probably caused by "eine Schädigung der Wasserresorption, indes die Resorption gelöster Bestandteile vor sich geht." With the fishes mentioned, and with amphibia this explanation might also assist us to understand the relation of the constituents of the urine and of the blood serum. It was important to know to what extent this retro-resorption did not take place with a quicker flow of urine, thus causing the constitution of the urine to become more and more equal to that of the albumen-free blood serum.

The urine of big specimens of Scyllium canicula was collected by a method which is usually applied in the Zoological Station at Naples. By a special shape of the canule it was possible to collect the urine of female animals. This made it certain that no sperm or mucus came into the urine, which accordingly was always clear and free from albumen in the experiments.

After a small part of the urine had been tested for albumen, the freezing point and the resistance were determined and then the percentage of nitrogen was determined by KJEHLDAHL's method. From the values found the urea concentration was calculated. Since diuretics have no effect on sharks I tried to increase the secretion of urine by the "piqûre". After urine had been collected for one or two days, I took the experimental animals from the aquarium for a moment and pricked them with a two-edged lancet in the medulla in the region of the posterior edges of the eye-sockets in the median line. Examination of dead animals proved that the vulneration had affected the lower part of the fourth ventricle or somewhat more towards the tail the medulla. In the most successful experiments the wound was on both sides and the greater part of the dorsal half of the medulla had been cut.

Put back into the aquarium immediately after being wounded the shark swam about in a somewhat restless and uncoordinated manner, besides a slight acceleration of respiration generally existed. These phenomena disappeared after 10—30 minutes; the animal could then in no way be distinguished from the healthy ones.

In the following table, containing a summary of the results of the experiments, $\angle$ means the freezing point, R the resistance, U the percentage of urea, while "prick" means that the animal was wounded in the above mentioned manner.

These experiments show:

a. The urine of Scyllium canicula has normally a smaller osmotic pressure than the blood serum. This is caused by the considerable difference between the percentage of urea of the serum and of the urine.

TABLE.

	Time in hours	Quantity cc	Δ° C.	R at 25.1° Ω	U %
experiment I					
urine	48	11	2.241	19 48	0.428
experiment II					
urine	48	10	2.206	20.19	0.38
blood serum	—	—	2.331	34.35	2.18
experiment III					
urine	48	13	2.162	25.48 (at 14°)	0.377
"prick"					
urine	24	10	—	—	—
„	48	25	2.306	26.6 (at 14°)	1.566
„	48	20	2.266	27.2 (at 14°)	1.392
blood serum	—	—	2.326	44.9 (14°)	2.02
experiment IV					
urine	48	9	2 186	20.21	0.48
"prick"					
urine	3	5	—	—	—
„	25	40	2.291	23.4	1.8705
blood serum	—	—	2.316	36.412	2.28
experiment V					
urine	48	8	2 251	20.23	0.712
"prick"					
urine	3	10	—	—	—
„	24	25	2.281	?	2.494
blood serum	—	—	2.336	34.7	2.61
experiment VI					
urine	48	13	2.226	21.608	?
"prick"					
urine	24	15	2.246	22.83	2.03
blood serum	—	—	2.291	36.4	2.33

b. By the described vulneration of the medulla the secretion of urea rose to about double or more.

c. Sugar could never be detected in the urine after the "prick".

d. The osmotic pressure of the urine rises with increased diuresis and hence approaches that of the blood serum.

e. The resistance of the urine for the electric current is increased with stronger diuresis.

f. From *d* and *e* it may already be surmised that the percentage of urea has risen with the diuresis. This is confirmed by the experiments. The percentage of urea rose to 3—5 times its original value.

If the filtration theory be true, then, as STARLING says: "Mit wachsender Absonderungsgeschwindigkeit (muss) ceteris paribus Beschaffenheit des Harns nach Zusammensetzung, Reaktion und osmotischem Druck immer mehr der des Blutserums minus Eiweisz sich nähern[1])."

The experiments here described afford in my opinion an unexpected confirmation of the accuracy of STARLING's thesis and so a probability for the accuracy of LUDWIG's explanation of the secretion of the kidneys.

Mathematics. — *"On the stable positions of equilibrium of floating parallelepipeda."* By Dr. P. BRANDSEN. (Communicated by Prof. D. J. KORTEWEG.

(Communicated in the meeting of October 30, 1909).

1. In the following paper will be communicated in short something about the stable conditions of equilibrium of floating homogeneous parallelopipeda.

Resting on the principle of LEJEUNE-DIRICHLET: "the position in which the potential energy is minimal is a stable position of equilibrium," we shall have to find positions where the common centre of gravity of the floating solid and the liquid is as low as possible.

From this (see APPELL, Traité de mécanique rationnelle, T. III, 1903, p. 180—218) the following construction indicated by DUPIN, can be deduced.

To find the position of equilibrium of a solid we have to determine a surface (G) which is the locus in the solid of the centres of gravity of the volumes of liquid, by the immersed parts according to the law of ARCHIMEDES (which centres of gravity coincide for a

[1]) Quoted from METZNER (l. c.) p. 242.

homogeneous solid with those of the immersed parts themselves): the normals out of the centre of gravity of the whole solid to this surface (G') then indicate possible positions of equilibrium. For homogeneous solids the only requirement for stability is that the chosen normal corresponds to a minimal distance of the centre of gravity to the surface (G'), i.e. that this distance is smaller than the smaller of the two principal radii of curvature of the surface (G') at the foot of the normal. For non-homogeneous solids the centre of gravity must moreover lie on the concave side of the surface (G'), which condition is fulfilled of itself for homogeneous solids.

It is worth mentioning, that the surface (G') is convex in all points, that for homogeneous solids the surface (G') may be determined for the floating as well as for the immersed part, whilst we must still bear in mind the following property: If the position of a floating body is given and if we determine the maximum and the minimum of the moments of inertia of the section of the surface of the liquid with the body with respect to lines through the centre of gravity of that section, then there exist the relations:

$$\varrho_1 = \frac{T_1}{V} \quad , \quad \varrho_2 = \frac{T_2}{V}$$

where ϱ_1, ϱ_2 are the principal radii of curvature of the surface (G') in the determined point, whilst V is the volume of the immersed part and T_1 and T_2 are the maximum and the minimum moments.

2. We commence with the solution of a question put by HUYGENS (Œuvres complètes, XI, p. 121 and 122) after the minimal ratio of the length of a right cylinder to the greatest dimension of its base, supposed to be everywhere convex, in order that the cylinder be able to float stably on any heavier liquid in such a way that its generatrices take a horizontal direction, if only the stability be assured with respect to displacements, where that direction is retained.

Let in fig. 1 the curve PLQ be the base everywhere convex of the right cylinder. For a definite specific weight ε with respect to the liquid (which we suppose $< \frac{1}{2}$) we have to introduce planes cutting base as well as upperplane and thus to determine the surface (G').

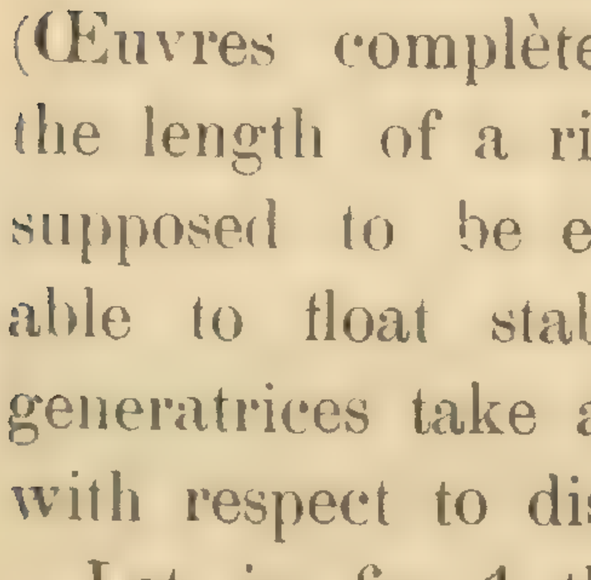


fig. 1

Fig. 1.

In this case where the generatrices are supposed parallel to the surface of the liquid we can determine the position of the corresponding principal centres of curvature of the surface (G') without knowing the surface (G). We then apply but the last mentioned property of § 1. If the centres found

lie in such a way that the centre of gravity of the cylinder lies between the centre of gravity of the immersed part and the centre of curvature which is the extremity of the smaller radius of curvature, a possible position of equilibrium will also be a stable one.

On the surface (G) lie the points obtained by determining the centres of gravity of the segments of the normal section, when every segment is equal to $\varepsilon \times$ the surface of the base and the normal section is taken halfway the height of the cylinder.

Let PLQ be such a segment and let R be its centre of gravity. The locus of these centres will be a closed curve, convex in all its points, because the surface (G) is convex. The centre of gravity G of the cylinder will lie inside this curve. Now there will be among the vectors of G to the points of the locus at least one minimum: for, starting from a definite vector we return to the same value the curve being closed. Let in fig. 1 GR be this vector or one of the other minimum ones. Now to this minimum vector corresponds a normal, pointing to a state of equilibrium stable for displacements where the parallelism of the generatrices with the surface of the liquid is retained, from which ensues that the extremity of the radius of curvature of the locus in R lies farther on this normal than G unless it coincides with G as a limiting case.

Let $PQ = \rho$ be the cord lying in the level plane. The latter is a rectangle with sides ρ and l (= length of the cylinder). The principal moments of inertia of this rectangle are:

$$\frac{1}{12}\rho l^3 \text{ and } \frac{1}{12}\rho^3 l.$$

The volume of the immersed part is il when i is the surface of the cut-off segment. The two principal radii of curvature in the point R of the surface (G) are therefore:

$$\varrho_1 = \frac{\rho l^2}{12\, i} \text{ and } \varrho_2 = \frac{\rho^3}{12\, i}.$$

Now ϱ_2 is the radius of curvature of the locus drawn in the figure. We have therefore, supposing RG to be a minimum vector, $\varrho_2 > RG$.

If now $\varrho_1 > \varrho_2$ or $l > \rho$ we are assured of the stability. The ratio $\frac{5}{3}$, mentioned by HUYGENS without proof and with reserve, is therefore unnecessarily large, it may be replaced by unity. For, the most unfavourable case is the one, where PQ, the level section belonging to the minimum value of RG, just coincides with the greatest dimension of the normal section. If then we take l larger than that greatest

dimension, l will certainly be larger than all other cords to be drawn, and therefore we shall have stability for all specific weights between 0 and $\frac{1}{2}$, so also between $\frac{1}{2}$ and 1. Hence we know that the ratio to be found cannot be larger than 1. If in fig. 1 $\varrho_2 = RS$, we can choose l still smaller, without endangering the stability, if only the extremity of ϱ_1 lies between G and S. Such a reduction would have to be possible in order to allow further reduction of the minimum ratio to be found for all specific weights for all convex sections.

Let us take a circle for the convex base of the cylinder. The loci of the centres of gravity of the cut-off segments are circles concentric with the given circle, i. o. w. in order to retain the notation of the preceding figure, we have $\varrho_2 = RG$.

If now $l = p = 2 \times$ the radius of the normal section then for $\varepsilon = \frac{1}{2}$ we have: $\varrho_1 = \varrho_2 = RG$, therefore $l : p$ cannot be reduced here without leading to $\varrho_1 < \varrho_2 = RG$. So we find here the right circle cylinder for $\varepsilon = \frac{1}{2}$ to be a limiting case, and we have given the proof that the minimum ratio to be found is unity.

3. In what follows we shall communicate the results of an investigation into the stable positions of homogeneous rectangular parallelepipeda. In the main we shall restrict ourselves to the communication of the method and of the obtained results, for the deduction of which we refer to our dissertation shortly to appear[1]). Prof. KORTEWEG investigated the positions completely when the parallelepipedon is long enough to float with its axis parallel to the surface of the liquid. (Nieuw Archief voor wiskunde, Second Series, eighth Volume, 1907, pages 1—25): Archives Néerl. (Sér. 2), Vol. XII, p. 362—388.

The following cases are imaginable if for convenience we suppose $\varepsilon > \frac{1}{2}$:

1st case: Four points lie above the liquid whilst the level section is a parallelogram or rectangle.

2nd case: One vertex lies above the liquid.

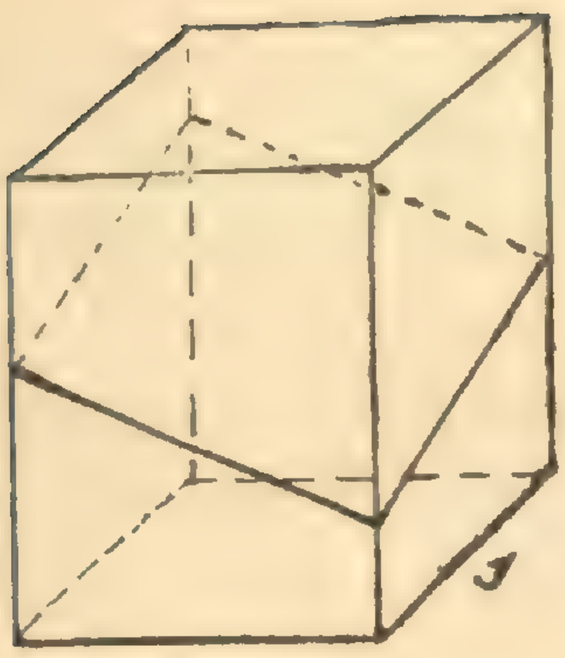
3rd case: Two vertices lie above the liquid. The level section is a trapezium, sometimes a rectangle.

4th case: Three vertices lie above the liquid. The level section is a pentagon.

5th case: Four vertices lie above the liquid. The level section is a hexagon.

These different cases are represented in the sketch below.

[1]) Has appeared since the Dutch paper was published.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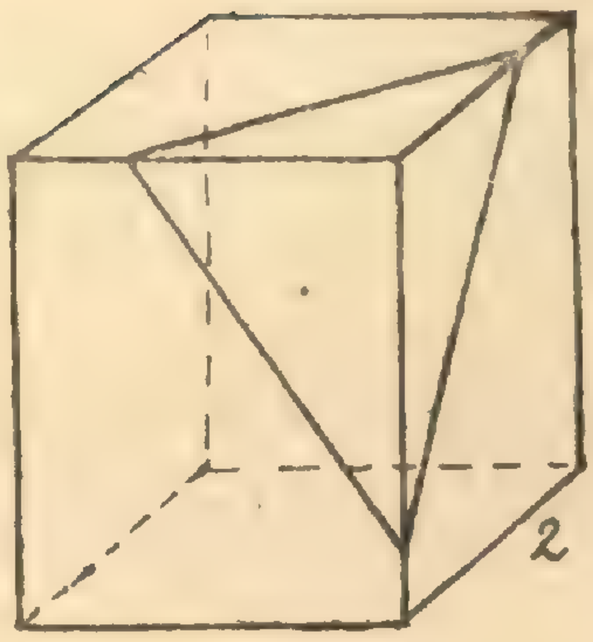
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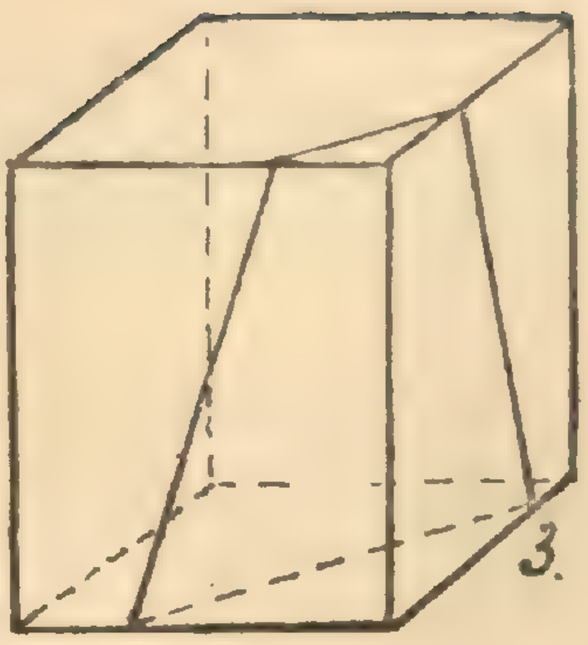
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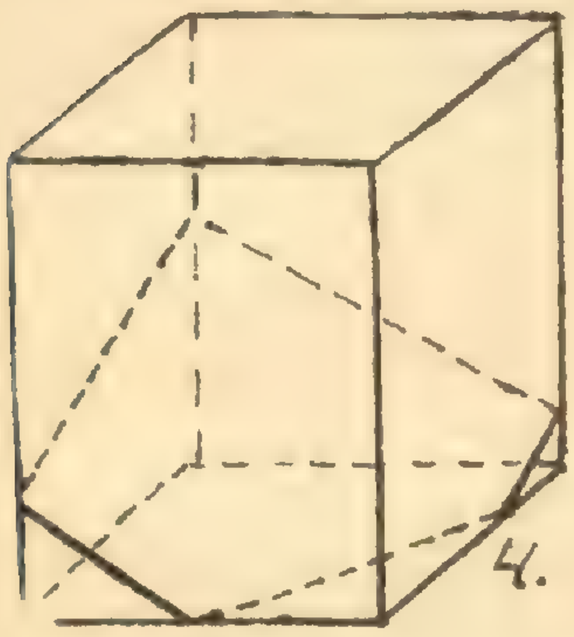
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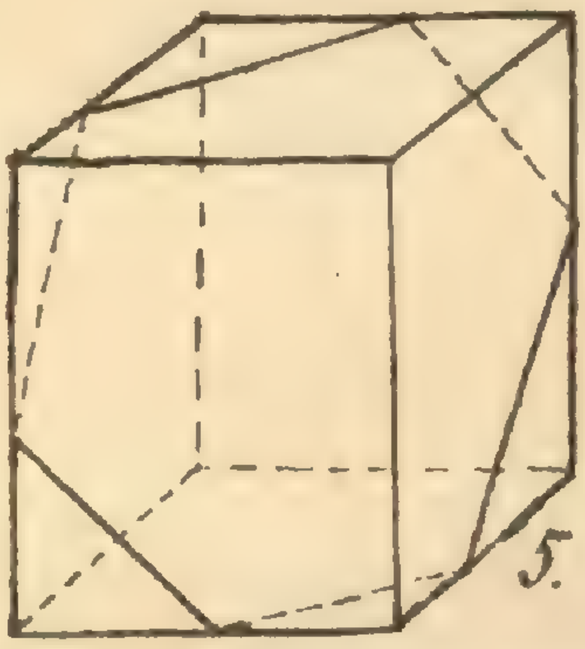
5.

For $\varepsilon < \frac{1}{2}$ the same figures can serve if we but exchange the immersed part for the floating one; in which case, however stability and equilibrium remain unchanged.

4. Following Prof. KORTEWEG's paper we shall make use of a graphic representation which will now however have to be a representation in three dimensions. Let us represent the lengths of the edges of the parallelepipedon by l, a and b, where $l > a > b$, then the positions of equilibrium will be determined by $\frac{a}{l} = \xi$, $\frac{b}{a} = \eta$ and ε (the specific weight of the solid with respect to the liquid on which it floats).

Let us now take a rectangular system of coordinates, in which ξ, η and ε are taken as coordinates, then each point in this system will denote a parallelepipedon of definite form and at the same time the value of ε. As now ξ, η, and ε are situated only between 0 and 1 we shall be able to represent the whole solution in the cube. This cube will be divided into compartments and to each of those compartments correspond one or more positions of equilibium. We have not succeeded in a complete division of that cube on account of the great analytical difficulties connected with it. What we found is summed up in the following.

5. *First Case.*

The surface (G) is an elliptic paraboloid in whose axis lies the centre of gravity of the parallelopipedon; the normals determining the positions of equilibrium lie therefore exclusively in the principal

planes of the paraboloid parallel to the planes of the parallelopipedon. Hence a pair of faces is always normal to the level of the liquid and from a closer investigation it appears that of the completely immersed edges the larger will run parallel to the level of the liquid. The complete solution is represented in the plate in fig. A. In the different compartments of the cube the positions are indicated by $(1l)$, $(2l)$, $(1a)$, $(2a)$, $(1b)$ and $(2b)$, according to the edges intersected by the surface of the liquid corresponding to l, a or b and according to their being normal: $(1l)$, $(1a)$, $(1b)$, or oblique: $(2l)$, $(2a)$, $(2b)$ with respect to the surface of the liquid.

Here follow the equations of the curved surfaces, separating the different regions:

Surf.	$OCDA$ and $O_1C_1D_1A_1$	: $6\varepsilon(1-\varepsilon) = \xi^2\eta^2$
„	$OCEA$	: $6(1-\varepsilon) - 8(1-\varepsilon)^2 = \xi^2\eta^2$
„	$O_1C_1E_1A_1$	: $6\varepsilon - 8\varepsilon^2 = \xi^2\eta^2$
Cylind. surf.	OG, OD and O_1G_1, O_1D_1	: $6\varepsilon(1-\varepsilon) = \eta^2$
„	OH, AE	: $6(1-\varepsilon) - 8(1-\varepsilon)^2 = \eta^2$
„	O_1H_1, A_1E_1	: $6\varepsilon - 8\varepsilon^2 = \eta^2$
„	GUG_1, DRD_1	: $6\varepsilon(1-\varepsilon)\eta^2 = 1$
„	HLS, ENT	: $\{6(1-\varepsilon) - 8(1-\varepsilon)^2\}\eta^2 = 1$
„	H_1L_1S, E_1N_1T	: $(6\varepsilon-8\varepsilon^2)\eta^2 = 1.$

6. *Method of the remaining cases.*

We place the parallelopipedon in such a manner, that 3 edges coincide with the axes of coordinates. We call the edges $2l$, $2a$, and $2b$, and the segments which the level plane or this plane produced cuts from the x, y and z-axis resp. u, v and w, which segments are always to be taken positively. Finally we call the coordinates of the partial centre of gravity (i. e. of immersed or floating part) x, y, z, which quantities are thus well-known functions of u, v and w different for the different cases.

As the specific weight is supposed to be given, the volume of the immersed or floating part is known, so that between u, v and w a relation exists:

$$F(u, v, w) = 0; \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

which again turns out differently for the different cases.

Now x, y, z is a point of the surface (G). The equation of surface (G) is therefore determined by elimination of u, v and w out of (1) and the three equations expressing x, y and z in u, v and w. Furthermore it is a property of the surface (G) that the normal in a point x, y, z is perpendicular to the corresponding level plane.

The equations of a normal are therefore:

$$u\,(X-x) = v\,(Y-y) = w\,(Z-z);$$

this normal must pass through the centre of gravity of the parallelopidon so that the condition of equilibrium becomes:

$$u\,(a-x) = v\,(b-y) = w\,(l-z) = \sigma, \quad . \quad . \quad . \quad . \quad . \quad (2)$$

where σ becomes known after the position of equilibrium being calculated.

To judge the stability it is necessary to know the position of the principal centres of curvature. On each normal are two of such points. They form together a two-sheeted surface which we shall call for the future surface (M). Let ξ, η and ζ be the coordinates of the point of (M) corresponding to an arbitrary point x, y, z of (G), then we find:

$$u\,(\xi - x) = v\,(\eta - y) = w\,(\zeta - z) = S, \quad . \quad . \quad . \quad . \quad (3)$$

where S is introduced for convenience. As now also the normal in one of the points $x + dx$, $y + dy$, $z + dz$ of the surface (G) must pass through ξ, η, and ζ on account of two adjacent normals intersecting each other in this point, this furnishes after substitution in (3):

$$\left(\xi - x - u\frac{\partial x}{\partial u}\right)du - u\frac{\partial x}{\partial v}dv - u\frac{\partial x}{\partial w}dw - dS = 0,$$

$$-v\frac{\partial y}{\partial u}du + \left(\eta - y - v\right)\frac{\partial y}{\partial v}dv - v\frac{\partial y}{\partial w}dw - dS = 0,$$

$$-w\frac{\partial z}{\partial u}du - w\frac{\partial z}{\partial v}dv + \left(\zeta - z - w\frac{\partial z}{\partial w}\right)dw - dS = 0.$$

Now the point $x + dx$, $y + dy$, $z + dz$ lay on the surface (G), therefore

$$\frac{\partial F}{\partial u}du + \frac{\partial F}{\partial v}dx + \frac{\partial F}{\partial w}dw = 0.$$

Eliminating out of the last four equations du, dv, dw, and dS, after having introduced $\xi - x = \frac{S}{u}$, $\eta - y = \frac{S}{v}$, and $\zeta - z = \frac{S}{w}$; then we find:

$$\begin{vmatrix} S - u^2\frac{\partial x}{\partial u} & -u^2\frac{\partial x}{\partial v} & -u^2\frac{\partial x}{\partial w} & u \\ -v^2\frac{\partial y}{\partial u} & S - v^2\frac{\partial y}{\partial v} & -v^2\frac{\partial y}{\partial w} & v \\ -w^2\frac{\partial z}{\partial u} & -w^2\frac{\partial z}{\partial v} & S - w^2\frac{\partial z}{\partial w} & w \\ \frac{\partial F}{\partial u} & \frac{\partial F}{\partial v} & \frac{\partial F}{\partial w} & 0 \end{vmatrix} = 0.$$

This determinant proves to be of the following form

$$S^2 - \psi S + \chi = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

Out of the properties of the (G) and (M) surfaces we know that (4) will be sure to have two positive roots corresponding to the two points of surface (M). Now the condition of stability is, that the centre of gravity of the parallelopipedon lies between x, y, z of the surface (G) and the most adjacent point ξ, η, ζ on the normal in x, y, z. Out of (2) and (3) then follows, that σ must be smaller than each of the roots of (4), which comes to:

$$\sigma < \tfrac{1}{2}\psi \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

$$\sigma^2 - \psi\sigma + \chi > 0. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

The calculations of the positions of equilibrium soon become impossible, but the shape of the surface (M) often makes it possible that much is said of the positions of equilibrium. This surface (M) is the locus of the points where 2 adjacent normals of surface (G) coincide. When passing the surface (M) we shall have to get 2 real normals more or less, whilst on the contrary the number of normals we can construct, out of 2 given points, to the surface (G) is the same if starting from one point we can reach the other without passing the surface (M), whilst their properties of being minimum distances or not remain: as these too change only after coincidence of two normals. We must thus try to determine the number and the shape of the compartments in which the surface (M) divides the space. The situation of the parts will inform us how the point out of which the normals are constructed lies with respect to the two sheets of the surface (M), which position is again the same for all points in the same compartment. To obtain then a graphic solution, the results found must be applied in the cube discussed in § 4. Each compartment in that cube points to a definite number of positions of equilibrium, amongst which again a definite number of stable positions appear. After this we must still investigate whether the normal indicating a stable position points to a serviceable level plane, i.e. to an immersed part of the form for which the surface (G) is determined.

In this or suchlike manner, the following results have been obtained.

7. *Second case.*

This is not to be realized: no parallelepipedon can float in a stable way with but one point above or below the surface of the liquid; yet the latter case, where $\varepsilon < \frac{1}{2}$, is submitted to a closer investigation on the supposition, that the faces of the parallelepipedon, concurring in the immersed point, are indefinitely produced without

possessing mass. The study of the surface of the centres of curvature of the surface $xyz =$ constant leads to a great number of possible ways of floating, about which however we refer to the dissertation.

8. *Third case.*

In as far as the floating edge is parallel to the level of the liquid, this case is solved in fig. B of the plate.

We sum up the equations of surfaces appearing in the cube, as far as $\varepsilon > \frac{1}{2}$.

Cylind. surf.	AE	$: \eta^2 = 6\,(1-\varepsilon) - 8\,(1-\varepsilon)^2$
,,	AF	$: (1+\eta)^{\frac{2}{3}} - (1-\eta)^{\frac{2}{3}} - \frac{4}{3}\{6\eta\,(1-\varepsilon)\}^{\frac{1}{3}} = 0$
,,	$EQNT$	$: \eta^2\,\{6\,(1-\varepsilon) - 8\,(1-\varepsilon)^2\} = 1$
Surface	$OAEC$	$: \xi^2\,\eta^2 = 6\,(1-\varepsilon) - 8\,(1-\varepsilon)^2$
,,	$OAFC$	$: \xi^2 = \frac{3\,\lambda-\lambda^2}{2}$, $\eta^2 = \frac{2}{3\,\nu-\nu^2}$, $\lambda\nu = 8\,(1-\varepsilon)$
,,	MEC	$: \xi = 4\eta\,(1-\varepsilon)$
,,	MER	$: 1 = 4\,\xi^2\,\eta\,(1-\varepsilon)$
,,	$EQNTRE$	$: \xi^2 = \frac{2}{3\,\lambda-\lambda^2}$, $\eta^2 = \frac{3\,\lambda-\lambda^2}{3\,\nu-\nu^2}$, $\lambda\nu = 8\,(1-\varepsilon)$;

where λ and ν are parameters.

The position where the longest edge is parallel to the level of the liquid, appears in (as far as $\varepsilon > \frac{1}{2}$):

1. The cylindrical region, of which ENT is base, from which the region $EQNTRE$ must however be excluded.

2. The cylindrical region with AEQ as base, from which must be excluded the part cut off from it by the surface MER.

In the cube this position is indicated by $(3l)$. The position, where the edge being the mean in length, is parallel to the level of the liquid, appears in the region between the surfaces $OAECO$ and $OAFCO$, from which we must separate the part cut off from it by surface MEW belonging to MEC.

In the cube this position is indicated by $(3a)$.

A stable floating with a shortest edge above the liquid proved impossible.

In the following a parallelepipedon is supposed with *square section*, so that when the edge of the square section is represented by $2a$ and the unequal edge by $2l$, the positions are perfectly determined by $\frac{a}{l} = \xi$ and ε. The third case, as far as no edges $2l$ are intersected by the level plane and the square section is not normal to the level-

plane, proves to be impossible for $a > l$. For $a < l$ it can however be realised, for values of ξ and ε (as far as $\varepsilon > \frac{1}{2}$), determined by the equations

$$\xi\zeta = \frac{2\zeta^2}{3+\mu^2} + \frac{1+\mu^2}{3+\mu^2} \text{ and } 24\xi^2\zeta^2(1-\varepsilon) = (3+\mu^2),$$

where ζ and μ may be the coordinates of a point inside the region ABC out of fig. 2. In this figure the equation of AB is: $1 + \mu^2 = 2\zeta^2$ and of BC: $\frac{2\zeta^2}{3+\mu^2} + \frac{1+\mu^2}{3+\mu^2} = \frac{1}{2}(1+\mu)$.

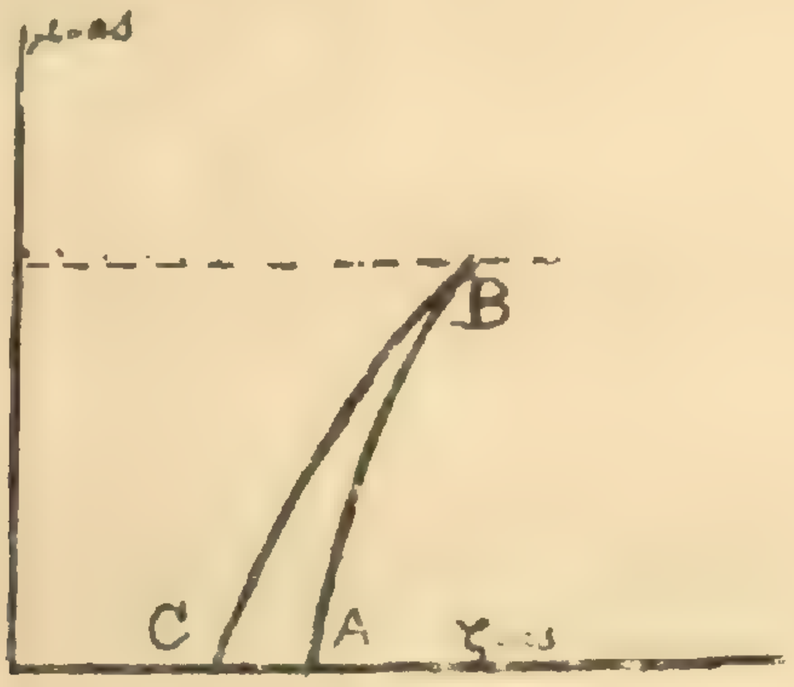

Fig. 2.

9. *The fourth case* was treated only for parallelepipeda with *square section*, where three vertices of the pentagonal level section were supposed to lie on the unequal edges $2l$. Only such positions that the level section is normal to one of the two diagonal planes passing through the unequal edges, proved to be possible. Such positions appear, in as far as $\varepsilon > \frac{1}{2}$, for values of ξ and ε determined by

$$\left.\begin{aligned} \frac{1}{\xi} &= \frac{w^4-2s^4}{2(w^3-2s^3)} + \frac{1}{w-s} - \frac{w^4-4ws^3+2s^4}{2(w-s)^2(w^3-2s^3)} \\ \frac{w^3-2s^3}{(w-s)^2} &= \frac{6(1-\varepsilon)}{\xi} \end{aligned}\right\}$$

where w and s are the coordinates of a point situated inside the region OQP in fig. 3.

The equations of the curves out of fig. 3 are:

Of PQ: $(w-s)^2(w^4-4ws^3+2s^4) = w^4-2w^3s+2s^4$,

Of OP: $w = 2s$

Of OQ: $3(w-s)(w^2-2s^2)(w^3-2s^3) =$

$(w^6-6w^4s^2+8w^3s^3-6w^2s^4+4s^6)(w-s)^2+(2w^6-6w^4s^2-2w^3s^3+12ws^5-4s^6)$.

10. *The fifth case* finally proved very unfit to be treated. Only for the floating *cube* could it be investigated a little more closely.

P. BRANDSEN. "On the stable positions of equilibrium of floating parallelopipeda."

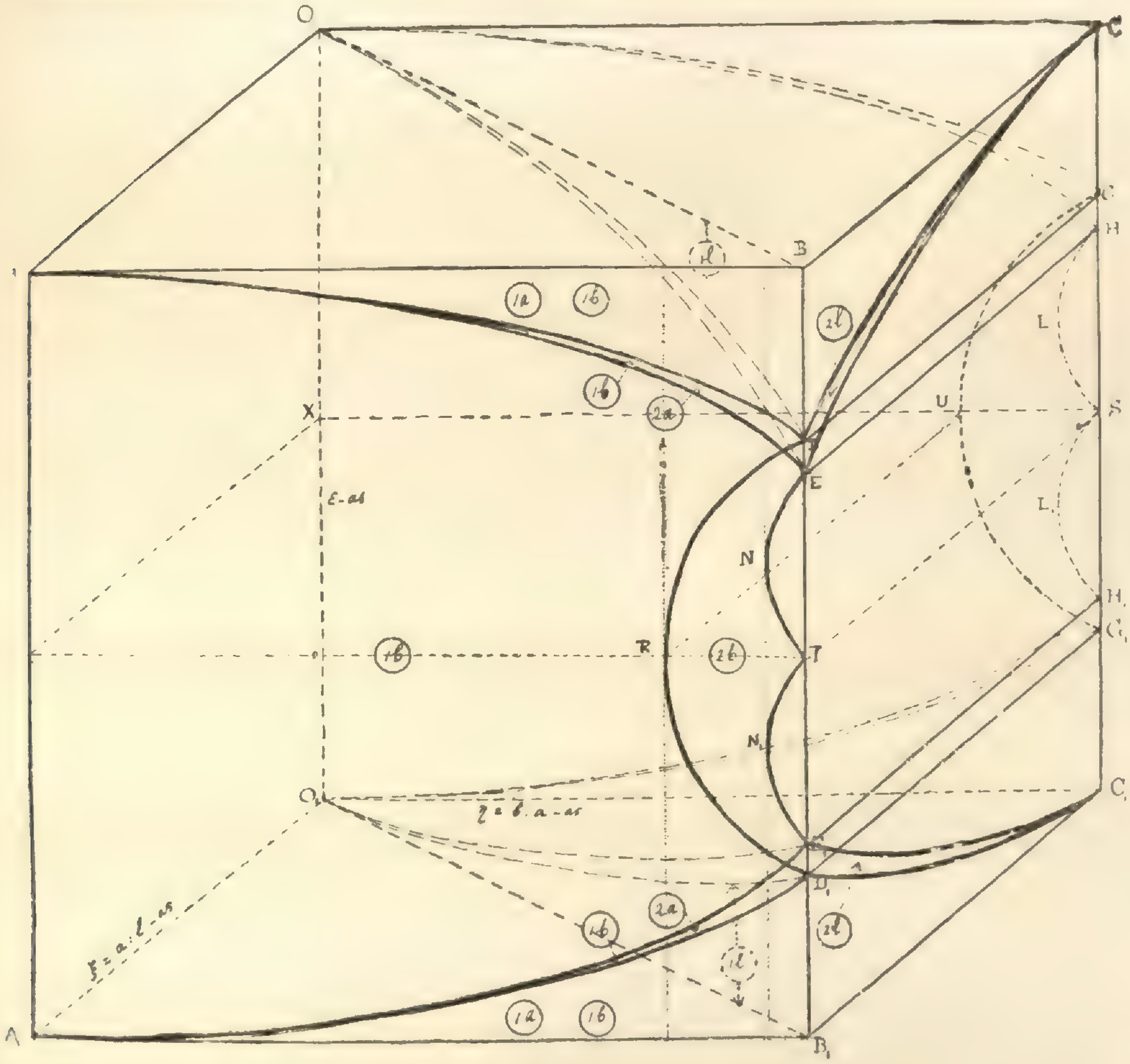

Fig. A.

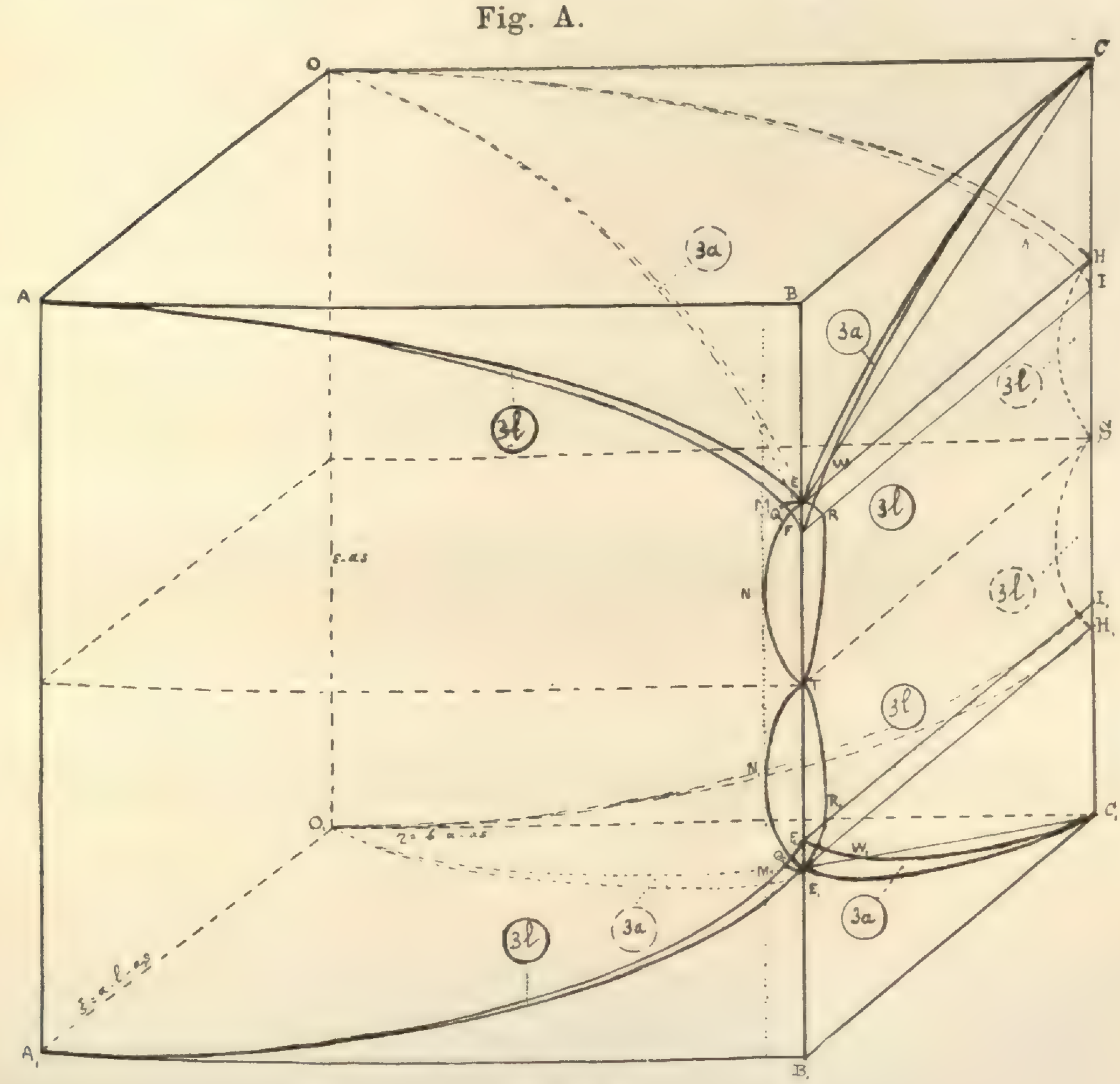

Fig. B.

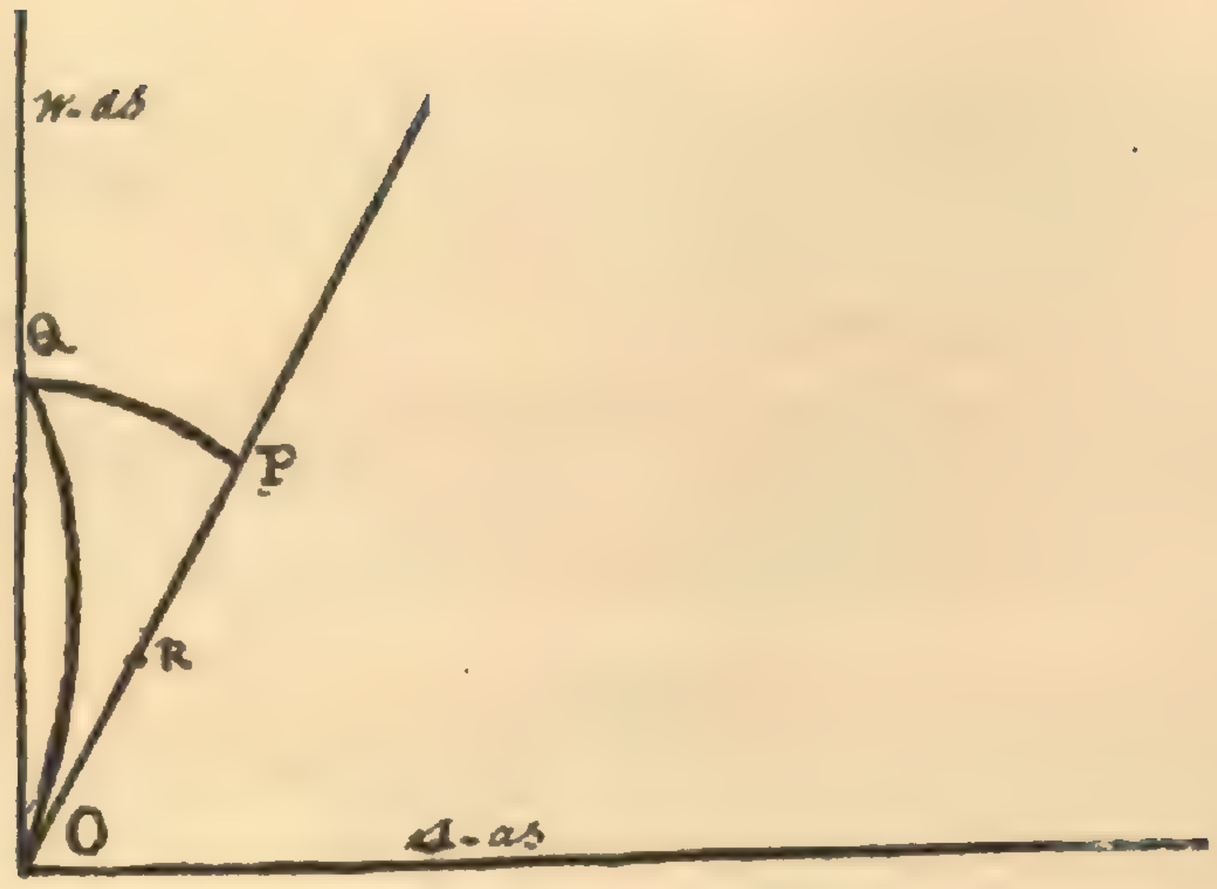

Fig. 3.

11. We conclude with a summary of the cases possible for a floating cube in so far as $\varepsilon > \frac{1}{2}$.

$1 > \varepsilon > 0.788 \ldots$	first case in vertical position.
$0.738 \ldots > \varepsilon > 0.75$	„ „ „ oblique „
$0.833 \ldots > \varepsilon > 0.77 \ldots$	fourth case, level plane a pentagon.
$0.833 \ldots > \varepsilon > 0.5$	fifth „ „ „ „ hexagon, where an interior diagonal is normal to the level of the liquid.

Moreover there are still possible positions belonging to the fifth case, where the interior diagonal is not perpendicular to the level of the liquid. The treatment of this is however attended with great difficulties.

Chemistry. — "*On partial racemism.*" By Mr. H. DUTILH. (Communicated by Prof. P. VAN ROMBURGH).

(Communicated in the meeting of October 30, 1909).

A recent article from FINDLAY and HICKMANS [1]) induces me to communicate the results of a research on the above subject, on which I have been engaged since August 1908, the publication of which has been delayed, however, through circumstances not under my control.

I have been occupied with the case of partial racemism which occurs in strychnine racemate and I have studied more in particular the behaviour of this substance in presence of its aqueous solution. Although this salt has already been subjected to an extensive

[1]) Journ. chem. Soc. Trans. **95**. 1386 (1909).

research under the same conditions by LADENBURG and DOCTOR [1]. I am of opinion that this research, although carried out fully and accurately, is attended with errors and that, therefore, the results do not represent the true state of affairs.

LADENBURG and DOCTOR have determined at different temperatures the solubilities of strychnine racemate and of strychnine *l*-tartrate and strychnine *d*-tartrate together, and they are of opinion that at the temperature at which the solubility of the partially racemic salt became equal to that of the mixture of its two component parts, the transition point of the strychnine racemate was situated, that is to say, that below that temperature the said salt would be stable in presence of its saturated solution but that at a higher temperature it would be resolved under those circumstances, with simultaneous separation of the least soluble of its dissociation products. By this process that temperature was found to be 30°, whereas by the tensimetric method 29°,5 was found. Moreover, the said authors have always endeavoured to determine the content in *d*- and *l*- strychnine tartrate of the solutions saturated with both salts by measuring the angle of rotation of those solutions and calculating the respective proportions from the known specific rotatory power of each of the two salts. Their observations led to the result that at the transition temperature the solution saturated with both components (these will in future be designated by *D* and *L* and their compound by *R*) would contain exactly 50% of both.

Immediately after their publication BAKHUIS ROOZEBOOM [2]) pointed out the improbability of their results and in the graphic representation given in his paper the way has been indicated how to set to work when studying the solubility phenomena in cases of partial racemism.

The two components in the system *L*, *D* and water, in which *R* can be formed from $D + L$, have, as we are dealing here with biactive substances whose configurations are partly each other's mirror images, at all temperatures a different solubility, which is shown by an asymmetric form of the solubility isotherms in this ternary system. This is elucidated by fig. 1. Suppose at a certain temperature the solubility of $L = OL$, of $D = OD$. If now *L* is dissolved in water already containing *D*, or *D* in solutions containing *L*, we obtain points situated on LC^1 or DC^1, respectively. C^1 represents the solution saturated with both *D* and *L*.

At the temperature of the transition point we obtain a similar

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[1]) Ber. d. d. chem. Ges. **32**, 50. (1899).

[2]) Zeitschr. f. physikal. Chem **28**, 494 (1899).

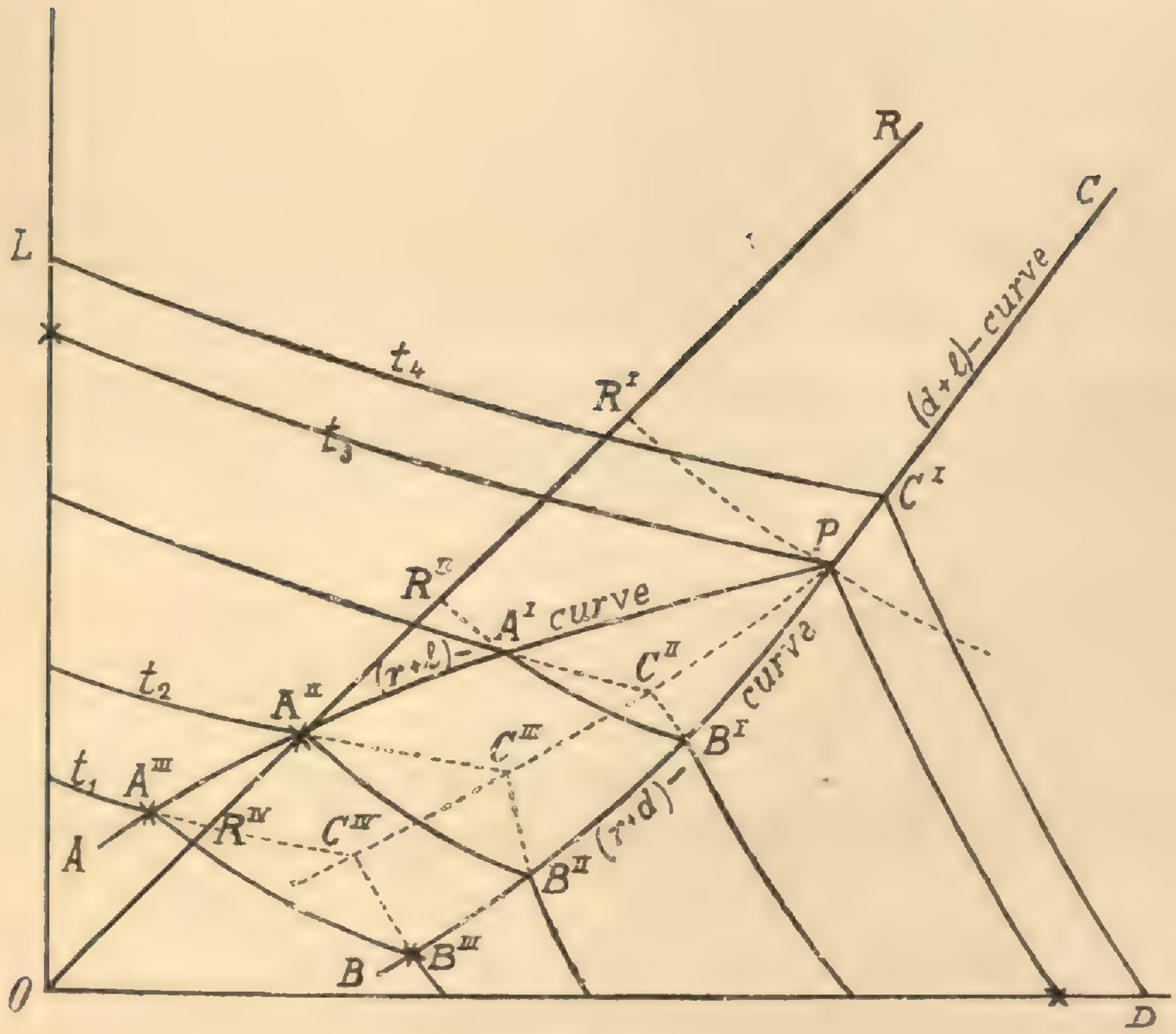

Fig. 1.

form of the L or D solubility curves, respectively, with this difference however, that a solution of the inner composition as represented by P can be also in equilibrium with R; in other words such a solution is obtained by shaking water with an excess of $R + D$, or $R + L$ or $L + D$. The zone of the partial racemate (R) in presence of solution is at this temperature limited to the point P. At a lower temperature this point extends to a line $A^I B^I$, $A^{II} B^{II}$, $A^{III} B^{III}$ etc. In the points A^I, A^{II} etc. the liquid is saturated with $L + R$, in the points B^I, B^{II} etc., however, with $D + R$. The points of the lines PA and PB, therefore, indicate the inner composition, as to D and L, of the solutions of $L + R$ and $D + R$ saturated at different temperatures.

The same also relates to the points on the line PC as regards the solutions saturated with $D + L$. The knowledge of the form of the lines PA, PB, and PC gives a complete insight into the stability phenomena of the double salt in presence of solution. Up to the temperature t_2 the partial racemate (R) can give solutions whose composition as to D and L is aequimolecular, above t_2 the substance begins to decompose with separation of the least soluble of its components (the line OR, which indicates the composition of R, then intersects the solubility line of L and so we obtain solutions

which are not saturated with respect to R, but to $L+R$) and this goes on when we increase the temperature, until above t_3 the salt R can absolutely exist no longer in the presence of any solution whatever (retardation phenomena excluded). The temperature region t_2-t_3 is called the transition interval.

In my research I have endeavoured to construct experimentally a similar diagram for the system strychnine *l*-tartrate, strychnine *d*-tartrate and water. For this purpose complete isotherms were determined successively by observing the change in the solubility of D, or L, if L or D was previously dissolved in water. In this manner I determined in this system the isotherm of 40° which, being situated above the transition point found by LADENBURG and DOCTOR, had a form which agrees very well with that of the lines LC^1 and DC^1. Isotherms at a lower temperature were then determined in a similar manner. First of all, I determined the D- and L-solubility curves at 25°, 16° and 7.5°, which, notwithstanding the fact that these are interrupted by the solubility curve of R, could be followed up to their intersecting point. The solubility as to $L+R$ and $D+R$ was then measured at the said temperatures (also that of $D+L$, and this always agreed very well with the intersecting point of the D- and L-solubility curves found. finally, by extrapolation).

When the solubility of these two salt couples was known, I placed them on the L- and the D-axis, connected these points with a straight line and looked, in the case of $L+R$ solutions, for the intersecting point with the L solubility curve; on the other hand in the case of $D+R$ solutions the intersecting point with the D curve was determined. In this manner, points on the lines PA and PB were determined successively in an indirect way and it was my intention to endeavour to trace the intersection of PA with OR, as the terminal point of the transition interval would then be known and the analogy between partial racemates and the inorganic double salts investigated by VAN 'T HOFF and his scholars be completely demonstrated. I succeeded in showing that at 7.5° the solubility of the racemate must be put down in the point where the L curve of the isotherm of that temperature intersects the line OR, and also that R and $R+L$ became equal as regards solubility. This gave the end of the transition interval and this temperature region, which in LADENBURG and DOCTOR's research was supposed not to exist at all, was shown to extend over about 20°. Saturated solutions of the partial racemate are therefore stable below 7.5° only; the solubility of the salt may,

however, also be determined at higher temperatures (by retardation in the decomposition) and points like R^I, R^{II} etc. may be thus established.

Similarly, the possibility of determining points like C^{II}, C^{III} etc. is due to the retardation in the formation of R from $D+L$ and the non-crystallisation of this salt from the solution.

The end of the transition interval is characterised by the fact that the saturated solutions of R and of $R+L$ are in equilibrium with each other; their vapour tensions are the same and, consequently, this point ought to be controllable by the tensimetric method (with the aid of BREMER-FROWEIN's tensimeter). The solubilities of R, D, and L at 7.5° have, however, become so small that the vapour tensions of these solutions do not practically differ from that of pure water at that temperature; consequently their mutual difference at a somewhat lower, or somewhat higher temperature and their equality at that temperature itself, is difficult to demonstrate. As a matter of fact I have not succeeded in verifying the result in this manner.

In the method followed by me I have carefully avoided analysing the liquids saturated with $R+D$, $R+L$ or $D+L$, by means of the polarimeter, as to their contents in L and D. For from the commencement of my research I was of opinion that such a method of analysis gives a wrong idea as to the inner composition of the solutions saturated with two salts.

A saturated $(R+D)$, $(R+L)$ or $(D+L)$ solution contains not only R and D, R and L, or D and L, but in those solutions it must be assumed that R- as well as D- and L-molecules will occur. Such solutions conform, being homogeneous phases, to the law of the mass-action and therefore an equilibrium is formed between D-, L- and R- molecules, which is determined not only by the temperature of the solutions, but also by their concentrations. This equilibrium may be expressed by $R \rightleftarrows D+L$.

LADENBURG and DOCTOR have determined the $(D+L)$ solubilities by pipetting off quantities of the saturated solution; these were then weighed and evaporated. After having ascertained in this manner the total concentration, the residue was dissolved up to a definite volume. After determining the angle of rotation α, the content in D and in L was calculated by means of the equation $\alpha = \alpha_d + \alpha_l$

$$\alpha = \frac{[\alpha_d]_D \cdot p_d \cdot S.l}{100} + \frac{[\alpha_l]_D \cdot p_l \cdot S.l}{100} \; ; \; \frac{100\alpha}{l.S} = [\alpha_d]_D \cdot p_d + [\alpha_l]_D \cdot (p - p_d)$$

in which $[\alpha_d]_D$ and $[\alpha_l]_D$ represent the specific rotatory power of D

and L, p the total concentration, p_d the content in D, p_l the content in L, S the sp. gr. of the solution and l the length of the tube.

This manner of calculation, however, is not admissible; for at a total concentration p and a content p_d for D, we have a content p_l for L and $p - p_d - p_l$ for R and the equation thus becomes:

$$\frac{100\alpha}{l.S} = [\alpha_d]_D \cdot p_d + [\alpha_l]_D \cdot p_l + [\alpha_r]_D \cdot (p - p_d - p_l).$$

In this are known α, $[\alpha_d]_D$, $[\alpha_l]_D$, $[\alpha_r]_D$, p, l and S.

The equation contains two unknown quantities and therefore we cannot get an insight into the inner composition of the solutions saturated with $(R + D)$, $(R + L)$ or $(D + L)$ by means of a polarimetric determination, even when assuming that the three compounds which occur in the solvent do not affect each other's rotations (which has not been ascertained).

To this it may be attributed that LADENBURG and DOCTOR found the point P, that is the transition point, on the line OR, whereas I have reasons to conclude that the isotherm of the temperature of the transition point, like all others, retains its asymmetric form.

This untrustworthy method of analysis has been applied by FINDLAY and HICKMANS in their investigation of the partial racemism of the *l*-menthyl-*r*-mandelic ester, which must be considered as the binary compound of the *l*-menthyl-*d*-mandelic ester with the *l*-menthyl-*l*-mandelic ester, a fact which has been clearly shown by the authors [1]) from the form of the melting point curve of those two esters. The partially racemic ester the binary system formed by possesses in this system a melting point curve of its own with a flat top and has, therefore, been partly resolved in the fused mass into the two components.

In their second publication [2]) the solubility isotherms are given at 35°, 25° and 10°. Of each isotherm only five points have been determined, namely, the solubilities of D, L, R, $R + D$ and $R + L$, whilst the solutions $(R + D)$ and $(R + L)$ have been analysed according to the above mentioned method.

It now appears to me that their graphic representation, which I reproduce, shows at once the deceiving conclusions drawn from the polarimetric method. If we look at fig. 2 we observe that the solubility of the *d*-acid ester decreases on addition of the *l*-acid ester to the solution at 10°, whilst it increases at 25° and 35°.

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[1]) FINDLAY & HICKMANS Journ. chem. Soc. Trans. **91**, 905, (1907).

[2]) Ibid. **95**, 1386 (1909).

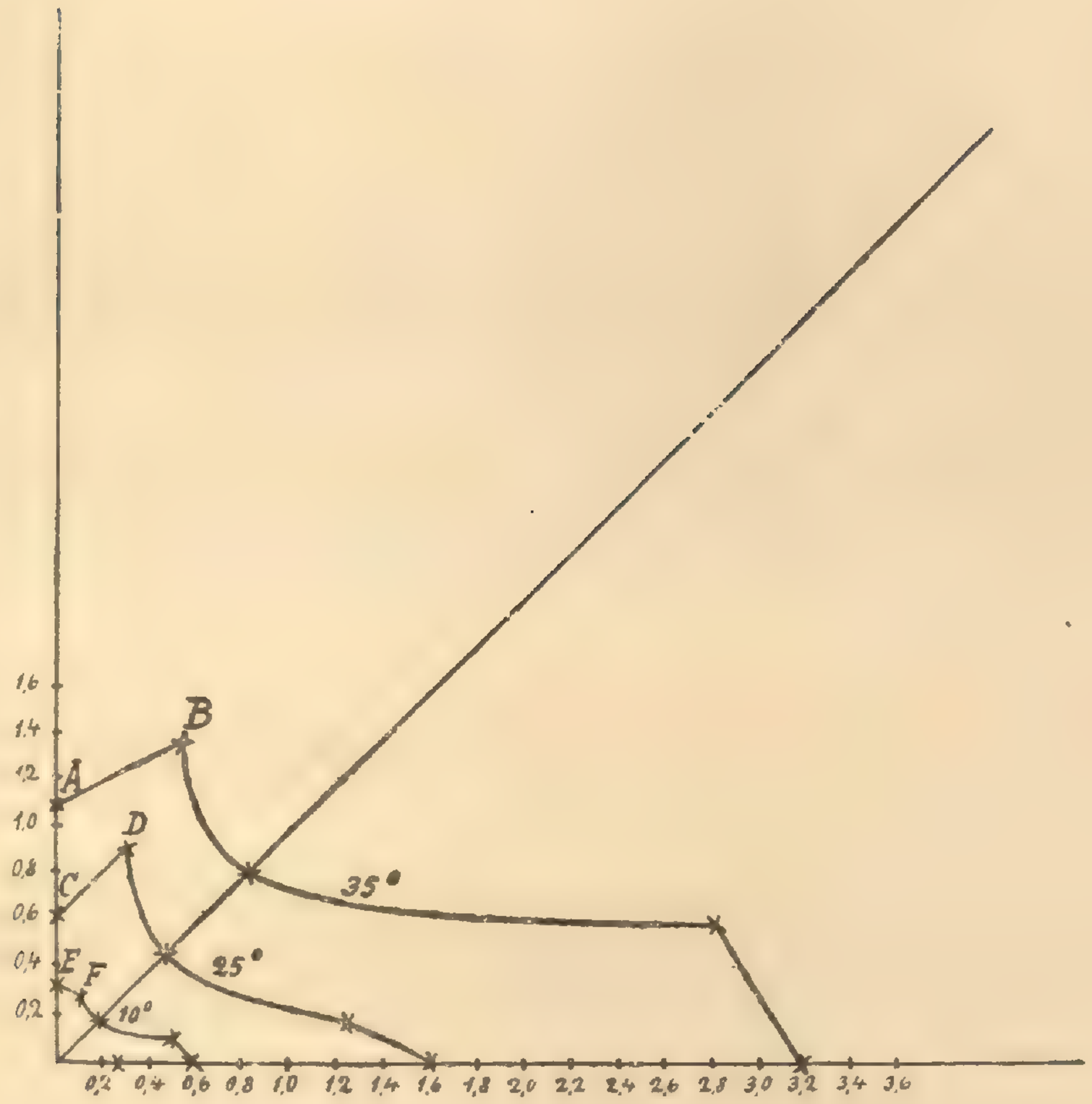

Fig. 2.

This will be noticed by comparing the lines *AB*, *CD* and *EF*. Such a behaviour is highly improbable, for it may be expected that at 35°, 25°, and 10° the effect on the solubility of the *d*-mandelic ester by the presence of the *l*-mandelic ester in the solution will manifest itself in an equal sense and not cause an increase of solubility at the one temperature and a decrease at the other. In the diagram of FINDLAY and HICKMANS, however, the course of the curves *AB*, *CD*, and *EF*, is, contrary to my investigations, determined exclusively by the situation of the points *B*, *D*, and *F*, whose position is deduced from an observed angle of rotation of the solution of the residues obtained by evaporation. It is not at all improbable that if these points were determined by another method their situation would be quite a different one. In this system the verification would have been a simple one, because the melting point curve of the binary system formed by the two biactive esters is known. The melting point for each content in *D*, or *L* in mixtures of $D + L$ is therefore known.

If now, FINDLAY and HICKMANS had only determined the melting point of their residues each time after evaporating their saturated solutions $(R + L)$ and $(R + D)$, they would have obtained from the situation of this melting point in connection with the melting point

curve of the binary system formed by *D* and *L*, so to speak "thermic" analyses of their solutions, which might have been compared with the polarimetric ones. It is to be regretted that these investigators have not applied this check as now it cannot, provisionally, be decided whether their isotherms exhibit the true form or not. From my results with the strychnine tartrates, I think I may decide upon the latter view. The truth may, however, be revealed by a determination of the complete isotherms or, perhaps with sufficient accuracy, by melting point determinations of the residues left on evaporation.

Utrecht. *Org. Chem. Lab. of the Univ.*

Botany. — "*Contribution to the knowledge of water-secretion in plants*". By Dr. W. BURCK.

(Communicated in the meeting of October 30, 1909.)

II. *Secretion of water in the flower.*

After TREUB [1]) showed in 1890 that the flower-buds of *Spathodea campanulata* Beauv. are filled with a watery fluid, a similar secretion of water in closed flower-buds has also been observed in a few other tropical plants.

LAGERHEIM [2]) observed the same phenomenon in a South-American Solanacea, *Jochroma macrocalyx* Benth. GREGOR KRAUS [3]) soon afterwards described the water-calyces of *Parmentiera cerifera* Seem., and HALLIER [4]) those of *Leea amabilis*. Later a detailed paper was published by KOORDERS [5]) on the flower-buds of the Bignoniaceae: *Parmentiera cerifera* Seem., *Crescentia Cujete* Linn., *Kigelia pinnata* DC., *Heterophragma adenophyllum*, Seem., *Stereospermum hypostictum* Miq.; of the Solanaceae: *Juanulloa parasitica* Ruiz et Pav., *Nicandra physaloides* Gärtn.; of the Verbenacea *Clerodendron Minahassae* T. et B. and of a Scrophularinea *Illysanthes* spec. Afterwards there followed important contributions by SHIBATA [6])

[1]) TREUB, M. Annales du Jardin botanique de Buitenzorg. Vol. VIII. 1890

[2]) LAGERHEIM. Zur Biologie der *Jochroma macrocalyx* Benth. Berichte der deutsch. bot. Gesellsch. Bd. 9, Jahrg. 1891.

[3]) KRAUS, G. Wasserhaltige Kelche bei *Parmentiera cerifera* Seem. Flora Bd. 81, p. 435—437.

[4]) HALLIER, H. Ueber Leea amabilis und ihre Wasserkelche. Annales du Jardin botanique de Buitenzorg. Vol. XIV. 1897.

[5]) KOORDERS, S. H.. Ueber die Blütenknospen—Hydathoden einiger tropischen Pflanzen. Annales du Jardin botanique de Buitenzorg. Vol. XIV. 1897.

[6]) SHIBATA, K. Zur Kenntnis der Kelch- und Knospenhydathoden. Bot. Centralblatt 1900. Bd. LXXXIII.

on the secretion of water in *Tecoma grandiflora* Loisel and *Katalpa Kaempferi* S. et Z. and on the Verbenaceae: *Clerodendron trichotomum* Thumb. and *Cl. squamatum* Vahl and by NILS SVEDELIUS [1]) on the water-calyces of tropical Convolvulaceae especially in *Stictocardia tiliaefolia* (Choisy) Hallier and *Operculina Turpethum* (L.) Peter.

Amongst other results these investigations have shown:

1. that glands which secrete the water, often at first produce mucilage, as was also found to be the case with the epidermal glands of the leaves, and that only in a single case i. e. in *Tecoma grandiflora*, the calyx-water reduced FEHLING's solution.

2. that the secretion of water on the inner surface of the calyx can begin at widely different stages of development of the flower: in many of the plants mentioned it begins very early, when the corolla and the sexual organs are first laid down, and ends towards the flowering period, but in *Jochroma macrocalyx*, *Clerodendron Minahassae* and *Juanulloa parasitica* the secretion of water continues during the time of flowering even up to the formation of the fruit. SVEDELIUS afterwards showed that in the above-mentioned Convolvulaceae the real secretion of water only begins after fertilisation, when the calyx-leaves continue to grow with the fruit.

3. that trichomes, of the same structure as those which secrete water on the inner surface of the calyx, may also be found elsewhere in the flower, especially on the outer surface of the calyx and the inner- and outer surfaces of the corolla.

This last point is especially important in interpreting the phenomenon.

KOORDERS mentions in this connexion, that such trichomes occur in large numbers on the outside of the calyx in *Nicandra physaloides*, *Kigelia pinnata*, *Stereospermum hypostictum* and *Clerodendron Minahassae*. They are laid down at a very early age, earlier than those of the inside, but often they already begin to die off, when those of the inner surface have scarcely begun to secrete water.

SHIBATA made the same observation with regard to the flowers of *Tecoma grandiflora* and SVEDELIUS observed also in *Stictocardia tiliaefolia* that, at a very early stage, the outside of the calyx is already covered with many small peltate glands.

Concerning the glands on the corolla, TREUB mentions that in *Spathodea campanulata*, the cushion-shaped trichomes of the inner surface of the calyx are afterwards reduplicated on the inside of the corolla, when the flower-buds have already undergone considerable development; KOORDERS found similar corollar glands in most of the

[1]) NILS SVEDELIUS. Ueber das postflorale Wachstum der Kelchblätter einiger Convolvulaceen. Flora, Bd. 96. Jahrg. 1906.

plants examined by him. In *Stereospermum hypostictum* Miq. he found them on the inside of the corolla: in other plants, viz. *Clerodendron Minahassae* T. et B., *Heterophragma adenophyllum* Seem., *Nicandra physaloides* Gärtn. and *Juanulloa parasitica* Ruiz et Pav. only on the outside and in *Parmentiera cerifera* Seem., *Crescentia Cujete* Linn. and *Kigelia pinnata* DC. both on the inside and on the outside.

In other respects but little is known concerning these epidermal trichomes. Whether the glands which cover the outside of the calyx secrete water, has not been further investigated. As regards the corollar glands, KOORDERS has indeed put forward the suggestion that they may participate in the production of the calyx-water and that in the older flowerbuds the function of secreting water may pass from the glands of the calyx to those of the corolla, but no definite conclusion was reached on this point; in any case the function of glands on the outside of the corolla had to remain hypothetical.

Experience shows that, as was already shown above, there is not only a close agreement in external shape and anatomical structure between epidermal waterglands and mucilage- and resinglands, but also between trichomes [1]) which secrete water and those secreting nectar; hence the mere presence of glands on the corolla does not enable us to infer the nature of these glands.

It is nevertheless quite possible that, in some of the plants examined by KOORDERS, the glands on the outside of the corolla secrete water during the bud-stage.

I am the more ready to assume this, because I am acquainted with a plant, in which, as will immediately be seen, the whole of the calyx water is derived from the corollar glands; for the rest this subject requires further investigation, because conditions may differ from plant to plant. As far as my experience extends to corollar glands in plants with and without water-calyces, these glands are not always water-glands and in those cases, where they really are,

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[1]) For the sake of brevity I only refer to what has been observed by HABERLANDT concerning the similarity of structure and shape of water- and nectar-secreting trichomes in connection with the stipular nectaries of *Vicia sepium* and the trichome- hydathodes on the upper surface of the leaflets; further I refer to the observations of KOORDERS concerning the so-called extranuptial nectaries on the calyx of *Kigelia*, *Crescentia*, *Parmentiera* and *Stereospermum* compared with the water-glands of these plants and to the observations of SCHWENDT on the extrafloral nectaries of *Muehlenbeckia sagittifolia* and various species of *Polygonum*, *Gossypium brasiliense* and other species of *Gossypium*, *Tecoma radicans* and *Ligustrum Regelianum*. (in Beihefte zum bot. Centralblatt Bd. XXII Heft 3. 1907.)

the secretion may also take place later, when the flower has emerged from the bud-stage.

This, for instance, is the case in *Nicandra physaloides*, to which further reference will be made.

In *Malva silvestris* and in *Sidalcea candida*, where numerous glands occur on the margin and on the lower surface of the petals, I have only observed the production of mucilage. In *Hibiscus esculentus* there is secreted, in addition to mucilage, glucose from the apical cell of the corollar glands; as we shall see, this case of glucose secretion does not stand by itself.

I now wish to show, by means of a few examples, that the trichomes on the outside of the calyx are most probably always real glands. and that therefore in plants with water-calyces water, or mucilage, or both, may not only be secreted on the inside of the calyx, but also in other parts of the flower. Now investigation has shown. that we must not consider this a peculiarity distinguishing plants with water-calyces from other plants; we must remember that generally, in many plants, the same glands occur in the so-called floral region as on the vegetative parts and that secretory trichomes are sometimes found on the axis of the inflorescence, on the bracts or on the peduncle, on the outer or inner surface of the calyx or corolla and not uncommonly on the sexual organs themselves.

I already pointed out above that on the young flowerbuds of *Fuchsia* exactly the same thin-walled, more or less club-shaped trichomes occur as may be found on the surface of the youngest leaves, and that secretion of watery mucilage also takes place on the surface of the flowers, when a cut plant is placed in a space saturated with aqueous vapour. What has here been said of *Fuchsia*, applies also to other plants; sometimes the secretion of water, and sometimes that of mucilage predominates, although the water-secretion generally occurs in glands which originally produced mucilage.

For the following examples I have selected by preference those which refer to plants with so-called water-calyces; as will be seen, they are not limited to the tropics. I first however, give an example of a plant, in which nectar-secretion, mucilage-secretion and water-secretion can all three be observed, and in which (as I have not often found to be the case) mucilage and water are secreted by different glands.

Finally some observations on the secretion of water by stamens will be added.

Abutilon Darwinii Hook *f.*

The Malvaceae do not belong to the plants, which have water-calyces, although in many respects they resemble them.

Instead of water, honey is here secreted on the inner surface of the calyx. This secretion is especially copious in the species of *Abutilon*. If an adult flowerbud be enclosed overnight in a glass-box, a fairly large quantity of nectar will be found on the broad flat bottom of the calyx, when the flower has opened next morning. In the hanging flower the nectar runs down along the inner surface of the calyx. Without having previously satisfied ourselves, that we are here dealing with glucose, we might suppose that *Abutilon* belonged to the plants with water-calyces.

In 1879 Behrens [1]) gave a detailed description of this nectary at the bottom of the calyx in *Malvaceae.*

It consists of a tender-walled tissue of small, irregular cells, of which those which constitute the epidermis have grown out to a multicellular thread-like trichome to which Behrens gave the inappropriate name of "Sekretions-Papille". As the apical cells of these papillae secrete, they together constitute an extensive secretion area. Now Behrens observed that the outflow of nectar is preceded by production of mucilage in the apical cell, taking place with formation of collagen. It is remarkable that trichomes of exactly the same shape as Behrens' "Sekretions-Papillen", which compose the nectary, are found in different places, both in the flower and outside.

Nestler, who in his paper on the water-secretion of the leaves of *Malvaceae* [2]), mentioned the various trichomes which may be found in these plants, distinguishes them as "lang-gestielte, mehrzellige Drüsenhaare", but only found them in *Abutilon* in comparatively small numbers, probably because he did not examine the very youngest parts for the presence of these glands.

On the outside of the young calyx, which in addition bears a thick covering of long, pointed, stellate hairs (Büschelhaare, Sternhaare) they are found in relativeiy small numbers, but the outside of the corolla, the very young peduncle and petiole, the young stipules and the very youngest leaves are thickly covered by them.

Besides these long pointed hairs and "Sekretions-Papillen" the young calyx has an abundant covering of much smaller glands of elliptical shape, which consist of one or two stalk-cells and 4 secre-

[1]) Behrens, W. J. Die Nectariën der Blüthen. Regensburg. 1879.

[2]) Nestler, A. Die Ausscheidung von Wassertropfen an den Blättern der Malvaceen und anderer Pflanzen. Sitzungsber. der Kais. Akad. der Wissenschaften Band CVI, Heft 1, S. 387.

tory gland-cells. These are the "kurze, mehrzellige blasig-kopfig gestaltete Haare" of NESTLER's paper.

If now, in order to convince ourselves of the secretory activity of these various glands we place a cut branch, bearing young flowers as well as young leaves, under a glass bell-jar kept moist, we can observe in a few hours that practically every "Sekretions Papille" of the peduncles, petioles, stipules and youngest leaves bears at its apical cell a large drop of mucilage, clear as a crystal, while the leaves are further covered with a watery layer of mucilage. The older full-grown leaves are almost dry on their upper surfaces, while their lower surfaces are covered with large drops, as already described by NESTLER.

From the young flowers under the bell-jar heavy drops soon hang down, and are replaced when they are removed by filter-paper.

The liquid does not contain glucose, nor is it mucilaginous. It is water, which is secreted by the smaller glands of elliptical shape, which behave from the beginning like true water-glands. Even there, where these glands occur on the very youngest leaves and flower-buds, they never form subcuticular mucilage or resinous products. The outer wall of the cells is always very thin; the cuticle is never seen to be raised by a substance formed inside the wall.

Three kinds of glands can therefore be distinguished in *Abutilon Darwinii*:

1. multicellular, thread-like trichomes, the "Sekretions-Papillen" of BEHRENS, which form the nectary at the bottom of the calyx, and which at the beginning of flowering, secrete glucose with mucilage from the apical cell.

2. glands of exactly the same structure, which secrete mucilage and also a little water; these are placed on the outside of the calyx and of the corolla, on the peduncle and petiole, on the stipules and on the youngest leaves.

3. glands on the outside of the calyx and on the under surface of the leaves, which only secrete water.

In other *Malvaceae* the same kinds of glands are generally found, but with a different local distribution.

In *Malva silvestris* the waterglands predominate; the leaves of the epicalyx, the outside of the calyx proper and the peduncles are thickly covered with them.

In the moist space under the bell-jar the young flowerbuds next morning have their epicalyx and calyx covered with water. The secreted water gradually increases to such an extent, that it literally drips down from the flowers.

Calystegia sepium R.Br. (*Convolvulus sepium* L.)

Nils Svedelius made important observations on the secretion of water on the inner surface of the sepals of tropical *Convolvulaceae*, particularly *Stictocardia tiliaefolia* (Choisy) Hallier and *Operculina Turpethum* (L.) Peter. Similarly the inner surface of the sepals of our indigenous *Calystegia sepium* is always moist and sticky, both in the bud and during the flowering stage. It may readily be observed that the slimy liquid is secreted by cushion-shaped or peltate glands, which are destributed over the inside of the calyx in relatively large numbers. These glands are here only partially arranged in groups of 1—5; most of them are distributed more or less uniformly over the inner surface. They do not occur on the outer surface of the sepal, but they are found in fairly large numbers on the outside of the bracts, and to a less extent on the inside of the bracts, on the petioles and on the leaves.

Under ordinary conditions there is rarely found so much liquid in the flower-buds, that it can be pressed out in drops between the fingers; nevertheless I have been able to observe this a few times. I do not know whether mucilage or water is also secreted after fertilisation, during the period of the ripening of the fruit, as in *Stictocardia tiliaefolia* and *Operculina Turpethum*. *Calystegia sepium* gives the impression of an impure, hybrid plant, its power of sexual reproduction is evidently much weakened and fruit formation by self-fertilisation rarely takes place.

If a branch of *Calystegia sepium* is placed with the cut surface under water in a moist bell-jar the glands on the surface of the bracts, on the petiole and on the leaves soon begin to secrete water and after a few hours the upper and lower surfaces of the leaves are covered with numerous droplets of water, which soon increase in size. By the coalescing of these drops both sides of the young leaves are soon covered with a layer of water and in the terminal bud in which the leaves cover one another wholly or in part, they are as it were in a water-bath, together with the enclosed axillary flower-buds, which themselves also secrete water on the inside and outside of the leaves.

The glands which secrete the water, secrete mucilage in the bud, when they are still very young; in this respect, therefore, they agree with the glands of the inner surface of the calyx. They are the "Trichomzotten" of Hanstein, which afterwards become water-glands.

Datura Metel L., *D. Stramonium* L.

The water-calyces of some other Solanaceae have been described by LAGERHEIM and by KOORDERS viz. *Jochroma macrocalyx* Benth., *Nicandra physaloides* Gärtn, and *Juanulloa parasitica* Ruiz et Pav.; the very large flower-buds of *Solandra grandiflora* are also always filled with a considerable depth of water. Similarly it may be said of the genus *Datura* that the corolla, stamens and ovary are developed in a water-containing calyx. Although the secretion of water in the species of this genus is not so abundant as in *Solandra grandiflora* or even in *Nicandra physaloides*, still a not inconsiderable quantity of water may always be found in the buds, and also later, during the flowering-period. But, whereas in other Solanaceae the water is secreted on the inner surface of the calyx, we find in *Datura* the peculiar modification, that the glands cover the outside of the corolla, whereas the inner surface of the calyx, while having many stomata, bears neither glands nor other trichomes. Nor is the glandular outer surface of the corolla free from stomata. In external shape and in internal structure these glands agree fairly well with those which are found in other Solanaceae; the secreting part in the glands of *Datura* also consists of three layers, but generally the middle cell is divided vertically in two, so that the glands are spherical rather than elliptical. In *Datura Metel* these waterglands are hidden between numerous long pointed hairs consisting of three or four cells.

In other plants with water-calyces, the calyx shows a much more advanced growth and earlier development than the other parts of the flower, so that when the calyx already forms a fairly considerable space filled with water, the other floral organs only just begin to appear on the thalamus. In *Datura*, however, this is not the case.

In very young buds of 3 mm. the corolla has already a length of $1^1/_2$ mm. and at this stage the water-glands are first formed.

When the corolla has reached a length of 5 mm. it is covered from top to bottom, and especially on the teeth of the limb, with glands of various sizes in various stages of development.

The outside of the calyx of *Datura* bears glands of approximately the same structure as those on the outside of the corolla and the same applies to the peduncle.

Further the leaves are covered on both sides, from their earliest youth, with glands which only differ from the glands of the corolla in a subsidiary character i. e. the shape of the secreting part. In somewhat older leaves, which are already more or less unfolded, they are especially numerous and closely packed on both surfaces

of the basal half of the leaf. For the rest the outside of the calyx, as also the leaf and the peduncle are covered with long hairs, which, from a broad base, run out into a point, are generally composed of 4 cells and have a highly refractive content; these hairs are especially numerous on the upper surface and are more or less limited on the under surface to the course of the veins.

Now if a cut young plant is placed for a few hours in a moist space, numerous droplets are secreted on the leaf especially on the basal portion; by the running together of these drops the leaves are soon covered with a layer of water. Such waterdrops appear at the same time on the outside of the young flowerbuds. The secretion of water is only absent from the very youngest leaves of the bud, but instead a mucilage-secretion takes place from the walls of the glands.

The waterglands of *Datura* are therefore originally mucilage-glands (Trichomzotten) which afterwards secrete water.

The long, erect, conical trichomes do not take part in the secretion of water. Not infrequently, however, a clear drop of mucilage is found on some of these hairs, on the apical cell or on one of the other cells.

Nicandra physaloides Gärtn.

As was already shown by KOORDERS the water-secretion of *Nicandra physaloides* agrees with that of *Spathodea campanulata* and of other plants in as much as the water-secreting trichomes are here also found on the inner surface of the calyx, and the secretion already begins when the other floral organs have scarcely been laid down. The plants grown here do not secrete any less than at Buitenzorg: the young buds are generally completely filled with water.

KOORDERS has already mentioned, that club-like trichomes, similar to those secreting the water on the inner surface of the calyx, are also found in almost as great numbers on the outside of the calyx and of the corolla.

I now add, that the young leaves also, from the earliest bud-stages onwards, are covered on either side with these trichomes, and that, as in *Datura*, they are afterwards found especially in large numbers on the basal half of the leaf.

In *Nicandra* also they are laid down very early; the very young leaflets of the bud, of no greater length than 1 mm., are already thickly covered with them. Likewise they are found to be originally mucilage-glands.

Nicandra further agrees with *Datura* in this respect, that the leaves — but here only on their upper surface — bear large conical

hairs of 3-4-cells, with highly refractive content; the distribution of these hairs is not determined by the veins. They no longer occur on the adult leaves.

In a moist space *Nicandra physaloides* behaves exactly like *Datura*.

After a few hours the young flowers and leaves — except the very youngest leaves of the bud — are covered with a layer of water, while the conical hairs show themselves to be mucilage-glands. The mucilage-secretion is slight, however; small droplets only occur, on a few hairs. This is not only observed in a moist space, but also on young plants, cut off and placed in a glass of water, but not covered by a bell-jar. Finally we may also observe in the moist space that the glands on the lower surface of the corolla secrete water during the flowering period, as was mentioned above.

Dahlia spec. et var. div.

As long as the capitula of *Dahlia* are still in the bud, the youngest florets are covered by the inner bracts of the involucre. Generally we may easily observe at this stage, that the hanging capitula are moist below, and that drops of liquid, between the involucral bracts and the florets, are discernible through the translucent bracts and may be moved to and fro by the slight pressure of a lead-pencil. Not infrequently a drop of liquid may also be seen hanging down from the capitulum, especially when the sky is overcast.

The droplets consist of water.

This secretion of water, although not inconsiderable, is much greater after fertilisation. The capitulum, which had opened out during the flowering period, once more closes up, while the involucral bracts grow considerably. The capitulum after flowering assumes the shape of a hanging top.

At the apex of the top a drop of water can nearly always be observed during the whole period of the ripening of the fruits; after removal this drop is renewed, in proof that water is continually excreted somewhere in the capitulum.

Further investigation shows that all ovaries and all corollae in the capitulum are covered on the outside with long trichomes, consisting of a row of about 15 cells with dense content, and reminding one of the nectar-secreting glands of Malvaceae. These trichomes are the glands which secrete the water.

In *Dahlia* every floret is placed in the axil of a bract. This bract completely enclosing the flowerbud, does not bear glands or other trichomes and does not take part in the secretion of water.

The ray-florets, which are folded longitudinally in the bud in a peculiar manner, bear most water-glands on their veins, but for the rest the latter are distributed over the whole outer surface. These water-glands appear in much smaller numbers on the disc-florets and are limited on the corolla-tube to the junction of the narrow and wide portions. It cannot be determined at which stage of the bud the water-secretion begins. In very young capitula the drops escape observation, or may not yet be present. They cannot be observed until the outer whorl of the involucre curves outwards. During flowering the secretion of water cannot, of course, be observed on account of evaporation. It is not until after fertilisation, when the capitulum again contracts, and the involucral bracts grow out and again approach one another, that the secretion once more becomes evident.

Coreopsis tinctoria Nutt., *Coreopsis lanceolata* L. *Cosmos hybridus* Hort. [1])

In the genus *Coreopsis* and in *Cosmos hybridus* Hort., as in *Dahlia* the young capitula are enclosed and covered by the inner whorl of involucral bracts which are here generally light-brown. If the capitulum be opened at this stage, the florets in *Coreopsis* and in *Cosmos hybridus* are found to be in a bath of fluid as in *Dahlia*.

In *Dahlia* the involucral bracts are superposed in two rows, without coalescing, but in *Coreopsis* and this species of *Cosmos*, arranged in a single row, they close up more tightly with their edges. This closing makes the extrusion of a drop here much rarer than in *Dahlia*.

After fertilisation the capitulum does not close again in these plants and a secretion of fluid during the ripening of the fruit does not occur.

Further investigation shows that here also the corolla is covered with the same long trichomes composed of 15 cells, as in *Dahlia*, but in these plants the young ovaries are free from trichomes. It is very remarkable, however, that in *Coreopsis* and in *Cosmos hybridus* these trichomes do not secrete water, but a fluid which when heated with FEHLING's solution, gives a very marked reaction for glucose: in this respect therefore the secretion agrees with that found by SHIBATA in the calyx of *Tecoma grandiflora*.

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[1]) I doubt somewhat whether this plant has been correctly included in the genus *Cosmos*. It appears to me rather to be a *Coreopsis*.

I shall be able to show afterwards, that it is by no means a rare case that glands, which might superficially be regarded as water-glands, secrete nectar. The agreement between water- and sugar-secreting glands is by no means limited to their external shape and anatomical structure (see foot-note, p. 402) but extends further.

It may be remembered that HABERLANDT [1]) expressed the view that there might be a phylogenetic relationship between nectar-glands and water-glands. HABERLANDT supposes that water-glands are very much older and that in many cases nectaries may safely be assumed to have been formed from them. He starts from the supposition that the difference between water- and nectar-secreting trichomes does not so much lie in the difference in composition of the product (for the cell-content of water-secreting trichomes also occasionally gives a reaction for glucose) as in the dependence on the hydrostatic pressure of the water-conduct system in the case of the secretion from water-glands, which dependence would not exist in the case of nectaries.

The gradual evolution of nectaries from hydathodes was regarded as giving to the plant the advantage that the secretion became independent of the root-pressure. We have already seen in the first part of this communication that the basis of this theory is incorrect and that it is difficult to defend any longer the hypothesis of a phylogenetic connexion between water-glands and nectaries.

In returning from this digression to observations I must point out that the left and right side of the tongue-shaped ray-florets are folded over in the bud towards their middle and that they are covered with secretory trichomes especially on the veins.

In the tubular disc-florets they are found especially on the limb (*Coreopsis tinctoria*) or only on the lowest portion of the corollar tube (*Coreopsis laciniata* and *Cosmos hybridus*).

Finally I wish to observe that in *Dahlia* and in *Cosmos* the corolla of the tongue-shaped ray-florets is not only covered with these long, glucose-secreting glands, but in addition with short stalked club-like glandular hairs. I did not find these in the two species of *Coreopsis*. They are not coloured by dilute stains, whereas the other glands take up stains. In shape they agree with those, which according to SOLEREDER are common among Compositae. The character of the walls suggests the secretion of mucilage; probably they only function later during the flowering-period.

From analogy with what may be learnt from *Calystegia*, *Datura* and *Nicandra* it might be expected that the trichomes which secrete

[1]) HABERLANDT, l. c. p. 58.

water or glucose on the outside of the florets in the capitula of those Compositae, might also be found in other places.

This expectation has been realized. In *Cosmos hybridus* and also in *Dahlia* [1]) the leaves are already covered in very early youth by two kinds of trichomes, firstly with such as correspond structurally to the water- and glucose-secreting glands of the florets and secondly with longer (*Dahlia*) or shorter hairs (*Cosmos*), which from a broad base are continued to a conical point, and have a thick wall with a striped cuticle. The former consist of a row of 15—20 thin-walled cells, succeeding one another somewhat like a chain of pearls, which in the middle of the trichome are as long as broad but which gradually become longer towards the apex. These cells, especially those of the upper half of the trichome, are filled with dense granular protoplasm. They are readily stained by aniline-dyes (methylene blue, rosaniline violet). In *Dahlia* they are chiefly found above the median and lateral veins, pressed against the surface on both sides of the leaf. In *Cosmos hybridus*, where the leaf-segments are very narrow, they are also found in largest numbers on the upper surface of the leaves above the vein, but here and there also on the lower surface. The other hairs have a different shape in *Cosmos* and in *Dahlia*, but it is not always constant in different parts of the same plant.

In *Cosmos* those occurring on the lower surface and on the margin of the leaf are composed of 6 cells; they arise from a broad and occasionally multicellular base, are conically pointed and curved like a sickle; the content is more or less highly refractive and the cuticle, especially that of the apical cell, is striped.

In *Dahlia* sickle-shaped trichomes occur on the margin of the leaf-segments, but those on the upper and lower surface of the leaf are much longer, are not curved and are not restricted in their distribution to the veins; the cuticle of all cells is striped. These hairs are much more numerous on the lower surface of the leaf than on the upper.

It is easy to verify that in the very youngest leaves the glandular hairs secrete mucilage (resin or balsam). In a somewhat more advanced state of development the glands above the median and lateral veins are partly, as it were, surrounded by mucilage; the cells of the upper half of the trichome have not infrequently perished during the mucilage secretion.

With cut plants placed in a glass of water under a bell-jar we observe exactly the same phenomenon as in *Calystegia* and the Solanaceae.

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[1]) In *Coreopsis* I did not investigate this point.

The folded leaf-segments of both species are soon covered on their inner and outer surfaces with a layer of mucilagenous water, but the secretion is not so abundant as in the other plants. In this case I did not succeed in producing the formation of drops.

Hence the glandular hairs, which in the capitula of *Dahlia* secrete water and those of *Cosmos* glucose, secrete a watery mucilage when they occur on the leaves. In this mucilage I could not find glucose, either in *Dahlia* or in *Cosmos*.

Melandrium album Garcke, (*Lychnis vespertina* Sibth.).

The female form of *Melandrium album* may also be reckoned among plants with watercalyces, but here there is the peculiarity that the secreted water takes the place which elsewhere is occupied by the nectar.

If a completely closed, immature flowerbud of the female plant be opened, the claws of the petals, and the walls of the ovary and calyx are found to be covered with numerous drops.

On superficial inspection the flower might be considered very rich in nectar, but if these droplets are sucked up by a strip of filter paper in order to test the paper with FEHLING's solution for nectar on the slide, not a trace of glucose is found.

In the male flower the various enclosed parts are also covered with numerous drops of liquid, long before the bud is ready to open, but here the droplets consist of nectar, which is secreted from the fleshy ring at the base of the stamens.

In other respects the nectary of the female flower appears fully developed; it resembles that of *Melandrium rubrum*, both in shape and in size, but it does not produce nectar, and the tissue is free from glucose.

A comparison of the two sexual forms of *Melandrium album* further shows that the female calyx, which is a little larger than the male and is somewhat constricted towards the top, is thickly covered on its inner surface with two kinds of hairs i. e. with elongated hairs of 3—4 cells, which terminate in a rounded gland-cell and with very similar hairs, which do not bear a gland-cell and terminate in a pointed apical cell.

These two kinds of hairs are not found on the inner surface of the calyx-tube; the margin and the inside of the limb alone are covered with glandular hairs.

Similar hairs, but generally having a somewhat longer stalk, cover also the outside of the calyx in both sexual forms.

After fertilisation, when the corolla has fallen off, the young fruit

is enclosed by the calyx which keeps pace with its growth and one can then observe that during the ripening of the fruit the space between the calyx-wall and the ovary is filled with a layer of very sticky mucilage, derived from the glandular hairs, which during this stage are fully active.

As regards the water, which is found in the female bud and in the open flower, I feel justified in assuming that this is secreted by the other, pointed hairs of the inner surface of the calyx; I feel the more ready to assume this, because in *Melandrium rubrum* there are only found on the inside of the calyx the mucilage-secreting hairs and not the pointed ones. Now in the female flowers of the latter species the inside of the calyx is also covered at all stages of development with a large number of glandular hairs, which evidently already secrete mucilage in the young buds; the fruits are also surrounded by a layer of mucilage, but no water is secreted in the flowers during the flowering period, presumably owing to the above mentioned absence of the pointed hairs [1]).

I now wish to show by means of two examples, that the secretion of water in the flower is not dependent on the outer or inner surface of the calyx, nor on the outside of the corolla (*Datura*), but that it can also occur on the stamens. [2])

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1) It is possible that water is also secreted in the female flower of *Silene Otites* Sm. With regard to this plant SCHULTZ mentions, that in Germany the nectar-secretion of the female flower is almost always absent, and that of the male flower very often, but that in the Tyrol the male (and the hermaphrodite) flowers, like the female ones, secrete honey at the outside of the ring which unites the bases of the stamens.

According to SCHULTZ insects can easely reach the honey of the male flower on account of its small depth and fairly wide mouth, but in the female flower this is probably always quite impossible, because the calyx and the petals are closely applied to the ovary.

I shook 50 male and as many female flowers of this plant with distilled water and tested the water for glucose. I found that the male flowers are abundantly provided with nectar, but that the water from the female flowers had not taken up a trace of glucose.

I did not succeed in observing drops of fluid in the female flower and cannot say whether water is secreted here as in *Melandrium album*. It seems to me most probable that we are here dealing with a flower which is wholly devoid of nectar and of water; this is the more likely because the trichomes on the inside of the calyx, which might possibly secrete water, do not occur in *Silene Otites*

2) With regard to the occurrence of "Trichomzotten" on the surface of the ovary e.g. in species of *Lysimachia*, in *Ononis spinosa* and in *Verbascum Blattaria* I must refer for the sake of brevity to my previous communication. Proceedings of the meeting of Nov. 28, 1908.

Secretion of water by the stamens.

Verbascum. If an almost fully grown bud of *Verbascum* be carefully opened, a clear drop of fluid will certainly be found in it.

KERNER [1]), who observed this phenomenon in *Verbascum phoeniceum* and *V. Blattaria*, considered it a very peculiar method of nectar-secretion, in which extremely minute droplets of honey were supposed to exude from the stomata.

If the fluid is sucked up with filterpaper and tested with FEHLING's solution it is found to contain no trace of glucose.

I have observed this secretion in the closed flowerbud of all species of *Verbascum* which I have been able to examine namely: *V. phlomoides* L., *V. Thapsus* L., *V. thapsiforme* Schr., *V. Phoeniceum* L., *V. Blattaria* K. and a hybrid of *V. Phoeniceum*.

The water may be most readily observed in buds which are about to unfold: in young buds it escapes observation or it is not yet present.

The source of this water cannot be indicated with absolute certainty. KERNER's view, that it is secreted by the stomata of the corolla, is erroneous, because stomata do not occur in the corollar epidermis of *Verbascum*. Neither are glands or other trichomes on the corolla.

I think, however, that I am justified in assuming that the water is secreted by the long hairs which cover the stamens. These hairs are generally coloured and have a peculiar structure; they consist of a single club-shaped cell with a long stalk and a finely marked cuticle. DELPINO regarded these hairs as "Handhaben zum Anklammern der blütenbesuchenden Insekten", whereas KNUTH considered, that they serve in addition to enhance the "Augenfälligkeit" of the *Verbascum* inflorescence, at least in those cases where the colour of the hairs differs from that of the corolla.

I consider them to be water-glands.

With FEHLING's solution they only give a very faint reaction for glucose and often the presence of glucose cannot be demonstrated at all. Occasionally a vertical, or more rarely horizontal slit is found in the club-shaped end: this is not a constant character, however.

I do not mean by this, that KERNER is wrong in saying that honey may be found in the flower of *Verbascum*. This genus is indeed considered to be nectarless, but with the exception of *V. Blattaria* an abundant secretion of honey is found on the wall of the ovary, which is covered with a dense felt of branched hairs with very thick walls. The honey which is excreted by the ovary-wall, is

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[1]) KERNER VON MARILAUN. Pflanzenleben. Zweiter Band. Neuer Abdruck. S. 159.

sucked up by this covering and retained through capillarity, so that during the flowering the ovary of *Verbascum* may be said to be covered by a mantle saturated with glucose.

Tradescantia virginica L.

Exactly the same phenomenon of the constant occurrence of a clear drop of fluid in the flower-bud may be observed in *Tradescantia virginica*. Here the phenomenon is even more striking, because there can be no question of the secretion of honey in the *Tradescantia*-flower. Nectaries are not found in the whole order of *Commelinaceæ*.

On sucking up the fluid with a strip of filter-paper and warming the paper on the slide with a drop of FEHLING's solution, we find in this case also, that the fluid does not contain a trace of glucose.

Just as I pointed out the probability that the drop of water in *Verbascum* is secreted by the hairs with which the stamens are covered, so I have reason to believe in the case of *Tradescantia*. that the staminal hairs, so well-known in physiology, secrete fluid in their capacity of water-glands.

Before concluding this communication I consider it necessary to explain briefly why the expression "Hydathode" has only been used exceptionally. It may be remembered that HABERLANDT brings together under this expression all "Apparate" and places, where secretion of water in the liquid state takes place in the plant, no matter in which way the water is set free, whether by active or by passive cells.

HABERLANDT supposed that the whole water-secretion of the plant, both at the apex and the teeth of the leaf and superficially by external or internal glands, is especially intended to prevent an injection of the intercellular spaces, whenever the water which the roots take up from the soil has caused an increase of hydrostatic pressure in the vascular bundles; HABERLANDT evidently meant to indicate by the expression "hydathodes or water-ways" the ways by which the excess of water is again got rid of.

It has been pointed out above that this conception is incorrect and that the activity of the internal and external glands is not directly related to the bleeding-pressure. The expression could therefore only be retained for places of least resistance, which allow the water which the roots have pumped up, to pass out when transpiration is impeded; for all other places we should, for the sake of clearness, speak of glands.

For the places of least resistance MOLL [1]) has, however, already

[1]) MOLL, J. W., Untersuchungen über Tropfenausscheidung und Injection bei Blättern. Verslagen en Mededeelingen der Koninkl. Akad. v. Wetensch. te Amsterdam Dl. XV, 1880.

introduced into the literature the expression *Emissarium* or *outlet of water*, which more clearly indicates the notion than the word "*Hydathode* or *water way*".

It is thus evident from the second part of this communication on the secretion of water in the plant, that a secretion of liquid not only takes place in the leaves, but often also in the floral region and on various parts of the flower.

These observations place in a different light the well-known phenomenon of water-secretion in the flower-bud of plants with so-called water-calyces.

Formerly we considered, that the secretion of water in the flower-bud was to be regarded as a useful arrangement, arisen in the struggle for life by natural selection, in order to protect the enclosed parts of the flower against dessication.

We now see that it is related to the phenomenon of the secretion of liquid on the surface of the plant and that the presence of water in the calyx is to be explained thus, that the water secreted by the glands of calyx and corolla is less exposed to evaporation in the closed calyx and so can collect there.

In other words, the secretion of water in the flower-bud did not arise because it is useful to the plant, but it may nevertheless be of advantage to the plant in these cases where the enclosed parts of the flower or the young fruits are exposed to the danger of dessication.

Chemistry. — "*Contribution to the knowledge of catalytic phenomena*". By Prof. J. BOESEKEN. (Communicated by Prof. A. F. HOLLEMAN).

(Communicated in the meeting of October 30, 1909).

I.

Through the researches of Perrier (Thesis 1896 and Comt. rend. **116**, p. 1300) and of myself (Rec. 1900, p. 19) it has been shown that in the reaction of FRIEDEL and CRAFTS it is not the aromatic hydrocarbon but the chloride or the anhydride which is attacked first by the aluminium chloride, as in many cases additive products could be isolated. These are then converted by the benzene derivative. Afterwards I have pointed out (Proc. 1907, p. 613), that the formation of these intermediate products cannot, as such, serve to explain the

catalytic influence of aluminium chloride, because the potential fall of the reaction:

$$R\,.\,Cl + Al\,Cl_3 = RCl\,.\,AlCl_3$$

must necessarily be deducted from the potential fall of the entire reaction, so that one must still search for the cause of the velocity of these intermediate reactions.

The chlorides which form these additive products with difficulty, or not at all, such as $C\,Cl_4$, $CH\,Cl_3$, $S_2\,Cl_2$, $P\,Cl_3$ etc. are as a rule attacked with greater violence than those which form solid molecular compounds with the catalyst.

As a matter of fact, I have come to the conclusion, and have stated so previously on account of the conversion of yellow phosphorus into the red modification (l. c.) that the explanation of the catalytic action must be looked for in the dissociating influence which the catalyst exercises when nearing the molecules.

Whatever we may think of the cause of the permanent coherence of the molecules, it is certain that there exists between the atoms a certain contrast, which cannot be quite effaced because the atoms cannot annihilate each other.

This contrast appears to be dependent on circumstances; if these change, the contrast also changes and this causes a modification in the existing equilibrium. To these changes belongs the approach of other atoms or atom-groups. The action of positive catalysts may now be represented as being that of atoms or atom-groups which owing to their immediate proximity so modify the existing contrast that another equilibrium can be attained by the first molecule.

In the reaction of FRIEDEL and CRAFTS, I therefore, assume that in the first place the aluminium chloride loosens the cloride (perhaps also the double bonds in the benzine derivative, for it appears that $AlCl_3$ exercises a condensing influence on unsaturated solutions, the least on benzene itself as was to be expected), and that the reaction can then take its course in the usual manner.

There are in the literature many indications that aluminium chloride has a purily dissociating action: I will mention the following cases. Methyl (ethyl) perchloroformate is resolved into CO_2 and CCl_4 (C_2Cl_6) (HENTSCHEL J. pract. [2] **36** p. 100 and MÜLLER Ann. **258** p. 61). Pentachloroethane is decomposed into HCl and C_2Cl_4 MOUREYRAT Bull. [3] **19** p. 179), $CH_2Cl\,.\,CH_2Cl$ into HCl and C_2H_2 (Bull. [3] **19** p. 445). Propyl chloride yields HCl and propylene (MOUREYRAT Bull. [3] **21** p. 616) etc.

Sulphurylchloride is resolved into SO_2 and Cl_2 (RUFF Ber. 1902

p. 4453), Oxalyl chloride, on heating gently, into CO and $COCl_2$ (H. STAUDINGER Ber. 1909 p. 3566).

In order to test the above view more closely I have added to these examples a few others, my chief object being to demonstrate (so as to elucidate the action of the catalyst in the FRIEDEL and CRAFT's reaction) that, in the first place the chlorine atom is loosened. As a rule the chlorine atoms will leave the molecule as such in a few instances only; this may be demonstrated, for instance, by gently warming C_2Cl_6 with aluminium chloride and it is this cleavage which causes no doubt the very complex course of the reaction between perchloroethane and benzene.

Considering that the reaction might proceed more readily if during the decomposition stable chlorine compounds such as HCl could be formed, I have chosen the following four substances: trimethylacetyl chloride, dichloroacetylchloride, chloral and trichloroacetyl chloride, which all contain the more or less overloaded CO-group, so that the reaction could be watched by the evolution of the carbon monoxide.

It is certainly remarkable that, notwithstanding the excess of chlorine atoms, trichloroacetyl chloride is the most difficult to attack. A priori, there is not much to say as to the velocity of these reactions. The result of this research seems to point out that the reaction proceeds particularly smoothly in the case where H and Cl atoms are present next to two adjacent C atoms and that in the second place the overloading plays a role.

Of the three other substances the trimethylacetyl chloride is the one attacked most readily, whereas the two isomers dichloroacetyl chloride and chloral do not show large differences.

In all cases, however, the reaction takes place in such a manner that the movable Cl atom combines either with an H atom or a molecule residue, in which case a molecule of carbon monoxide is always evolved.

I. Trimethylacetyl chloride is resolved at 0° according to the equation:

$$(CH_3)_3CCOCl = HCl + CO + C_4H_8$$

the isobutene is nearly entirely polymerised.

II. Chloral on being heated gently is decomposed in two directions.

A $CCl_3COH = HCl + CO + CCl_2$ ± 70—75%

and the CCl_2 residue is polymerised to C_2Cl_4

B $CCl_3COH = CO + CCl_3H$

a slight resinification cannot be entirely prevented.

III. Dichloroacetyl chloride, on being heated gently, is also decomposed in two directions.

$$A \quad CCl_2HCOCl = CO + CCl_3H \pm 60\%$$

$$B \quad CCl_2HCOCl = CO + HCl + CCl_2$$

the latter being presumably polymerised to C_5Cl_{10}.

IV. Trichloroacethyl chloride, when distilled repeatedly with $AlCl_3$, is decomposed in one direction:

$$CCl_3COCl = CCl_4 + CO$$

The first part of the research, a contribution to the theory of the decomposing action of aluminium chloride was herewith attained.

II.

As regards our second object, namely, the finding of the point attacked in the decomposition, the different behaviour of chloral and dichloroacethyl chloride gives already some indications.

If we assume that one of the Cl-atoms of the chloral is detached we may expect it to combine with the movable H-atom to form HCl and this indeed takes place. If the CO group or the H-atom were the point attacked we ought to obtain chloroform which I have now been able to isolate in very small quantities only.

On the other hand, and for the same reason, the formation of chloroform from dichloroacethyl chloride is to be expected; it is now the movable chlorine atom which is detached first; CO and CCl_2H then follow immediately. Being the most unsaturated one the latter group will combine with the chlorine atom to chloroform, hydrogen chloride occurring as by-product. The actual course of the reaction is therefore quite in harmony with the assumption that primarily the movable chlorine atom is rendered active.

The action of aluminium chloride on trimethylacetyl chloride is too violent to decide anything as to the first stadium of the reaction.

[That the chlorine atom exercises again a preponderating influence on the instability of the molecule is shown from the behaviour of the otherwise quite analogous pinacolin; this simply unites with the catalyst without forming a trace of CO even at 100°].

I have, therefore, observed the action of concentrated H_2SO_4 in regard to trimethylacetyl chloride because the behaviour of this acid is in many cases analogous to that of $AlCl_3$ and also because it forms the same kind of additive products (Recueil 1905, p. 222).

In this investigation the reaction appeared to pass through the following phases :

$$\text{I} \quad (CH_3)_3\,C.COCl + H_2SO_4 = (CH_3)_3\,C.COSO_4H + HCl$$

$$\text{II} \quad (CH_3)_3\,CCOSO_4H = H_2SO_4 + CO + C_4H_8$$

in which the isobutene was again first polymerised and then dissolved on prolonged action of the acid, with formation of sulphonic acids (Bistrycki and Mauron Berichte **40** p. 4377) and sulphur dioxide.

The chlorine atom is, therefore, first detached by the H_2SO_4 and in an analogous manner we may expect the same from $AlCl_3$.

Sulphuric acid itself supplies an H-atom; on using $AlCl_3$ this hydrogen must be withdrawn from the acid-chloride so that the two phases noticed separately when sulphuric acid was employed as catalyst, coincide in this case.

[It may be pointed out here that a 20% "Oleum" acts almost as energetically as $AlCl_3$. It is very probable that the real catalyst in sulphuric acid is the SO_3 which in concentrated acid is not utterly paralysed by the H_2O present].

Although the expulsion of the chlorine could be shown in a single (see above) instance only, I think I have succeeded in demonstrating from the progressive change of the reactions and also by indirect means, that the chlorine atom is rendered more movable.

In the reaction of Friedel and Crafts we must imagine that besides the chloride, the benzene derivative, being an unsaturated compound, is also rendered active by aluminium chloride which undoubtedly will take place most readily when the chloride, catalyst and benzene derivative form one homogeneous phase. The significance of the so-called ferments of G. Gustavson (J. pr. Chem. [**2**] 68 p. 209—234) must be particularly looked for in the fact that these are *very* loose compounds in which the three kinds of molecules mentioned have arrived in each others, immediate proximity.

The fact that the rays of light introduce the halogens into the chain must, therefore, be explained by assuming that the double bonds of benzene are rendered less active by the chemical rays than the hydrogen atoms in the chain, whereas on the other hand the latter are not rendered quite so active by aluminium chloride.

The experimental details of this research will be published elsewhere.

Delft. Org. Chem. Lab. Techn. University.

Geology. — *"On jurassic fossils as rounded pebbles in North-Brabant and Limburg"*. By Dr. P. TESCH (communicated by Prof. G. A. F. MOLENGRAAFF).

(Communicated in the meeting of October 30, 1909).

It is now a year ago that I drew the attention to the fact that the so-called "Kieseloolithstufe" of the Lower Rhine basin, recognised as a separate formation in 1905 by German geologists, proves to be also represented in the territory of Northern-Limburg [1]). At that time especially the stratigraphical conditions of these fluviatile strata were discussed, this factor having proved to be of practical interest for the explorations of coal in these regions and the remarkable silicified fauna being found in these deposits was only mentioned in passing. After having finished this paper the collected material of fossils originating from the sands of this geological horizon has still greatly increased. This augmentation is owing, besides to the new borings executed since that time, to the discovery of a good finding-place in a clay-pit in the hills east of Reuver during this summer [2]), so that the collection now at my disposal is complete enough to enable us to draw some conclusions concerning the geological age of this silicified fauna and to compare these results with the opinions published by other authors about this subject.

So for the discussion of the stratigraphical and lithological composition of these strata referring to my former papers treating this subject and to the literature which is mentioned there, I have to study in the first place the results obtained by others when examining those fossils of neighbouring German places.

These silicified and rounded petrifactions are mentioned for the first time by Prof. Dr. J. POHLIG at Bonn in the "Verhandlungen des naturhistorischen Vereins der preuss. Rheinlande und Westfalen", 1883, second half, page 225. Here the following genera are enumerated, originating from the white coarse sands of *Duisdorf*, not far from Bonn:

Milioliden?	*Cardium*
Vioa	*Mytilus*
Astraea?	*Pholas*
Encrinus	*Purpura*

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[1]) P. TESCH. Der niederländische Boden und die Ablagerungen des Rheines und der Maas aus der jüngeren Tertiär- und der älteren Diluvialzeit. Amsterdam, 1908.

[2]) P. TESCH. De klei van Tegelen, een onderdeel der "Kieseloolithstufe". Tijdschrift van het Kon. Ned. Aardrijksk. Gen. Deel XXVI (1909), p. 573

Pentacrinus
Cidaris
Serpula
Terebella
Membranipora
Monticulipora
Ostrea
Spondylus
Pecten.
Turritella
Turbo
Dentalium

The author believes those genera to prove an upper-senonian age for these organisms. The next year H. von Dechen already suggested some doubt as to the correctness of this conclusion in "Erläuterungen zur geologischen Karte der Rheinprovinz und der Provinz Westfalen", second volume, 1884, page 626. Meanwhile it was only in 1897 that the incorrectness of this opinion was demonstrated convincingly. In this year C. Schlüter published in his paper: "Zur Heimatfrage jurassischer Geschiebe im Westgermanischen Tieflande" (Zeitschr. der deutschen geol. Gesellsch., Volume 49, 1897, page 486) the results of his examination of the petrifactions of *Duisdorf*, in consequence of which he expresses this opinion that this fauna originates from *upper-jurassic* strata. By this author the following fossils are mentioned, *all collected in the same gravel-pits of Duisdorf near Bonn*:

Thamnastraea (*Astraea*) *microconus Goldf.?*
Millericrinus horridus d'Orb. (*M. echinatus Schloth. sp.*) (many stems in different varieties).
Cidaris florigemma Phill. (spines).
Cidaris psammosa Mösch.? (fragment of spines).
Rhabdocidaris trispinata Quenst.? (fragment of spines).
Serpula (tubes).
Rhynchonella? (fragment).
Ostrea gregaria Sow.? (fragment).
Ostrea hastellata Schloth.? (fragment).
Ostrea pulligera-ascendens Quenst.? (fragment).
Exogyra reniformis Goldf.? (fragment).
Pecten vitreus Röm.? (printing).
Turritella jurassica Quenst.?

C. Schlüter believes that those fossils more specially must belong to the middle part of the *Oxford*-horizon and must originate from the "*Terrain à chailles*" ("*Sequanien inférieur*") of French Lorraine, southern Baden, upper-Alsace, northern Switzerland and the Jura-mountains.

It was not before the geological exploration of the Rhine-province by the "geologische Landesanstalt" of Berlin was begun that the same strata as those of Duisdorf were found elsewhere too and that the silicified fossils were collected also on other places. In his description of these fluviatile deposits in the Lower Rhine basin Dr. G. Fliegel enumerates the following petrifactions: [1])

Millericrinus horridus d'Orb.
Cidaris-spines.
Serpula-tubes.
Alectryonia.
Nerinea.
Trigonia costata Park.
Belemnites
Quenstedticeras Mariae d'Orb.

Then this geologist publishes as his opinion that the greater part may be derived from *Malm*-deposits but also some species indicate an origin from younger *Dogger*-deposits.

After this compendium of what the German places have yielded now, I can pass to the discussion of the fossils collected in the same strata in *North-Brabant* and *Limburg*.

Some small pebbles in which round, lenticular and oval figures of the size of $^1/_2$ to 1 m.m., distinctly distinguished from the round anorganic oölites, are very numerous, may be considered as silicified *foraminifera-limestones*. With Pohlig those fossils can be classified with reserve under the *Miliolidae*.

Little fragments of *Spongiae* are present from different places, usually further totally irrecognisable. A fragment from the boring 1 near Vlodrop may belong to *Chemidiastrum stellatum Goldf. sp.*, another fragment from the same place to *Ensiphonella Bronni Mst. sp.* From the other fragments nothing is to be said with any probability.

The *Anthozoa* are also represented by some specimina. All fragments are however very incomplete. A specimen from the clay-pit near Reuver probably belongs to *Thamnastraea prolifera Becker*, a fragment from the boring 5 near Leemhorst probably belongs to the genus *Favia*; the rest however is too small and too much damaged to enable us to identify them.

By far the most numerous fossils are, with the serpula tubes, the remains of *Crinoidea*, so that this group forms the most characteristic part of this silicified fauna. Most of the stems certainly belong to

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[1]) G. Fliegel. Pliocäne Quarzschotter in der Niederrheinischen Bucht. Jahrbuch der Königl. Preuss. Geol. Landesanstalt für 1907, p. 100.

species of *Apiocrinus* and *Millericrinus*; especially the sub-pentagonal and knobbed stems of *Millericrinus horridus d'Orb.* (= *M. echinatus Schloth.*) are rather well recognisable. Also thin and slender, smooth and round stems and free segments occur, which may be derived from allied species. The biggest stems measure 10 m.m. in diameter, the thinnest ones 3 or 4 m.m. and the greatest length amounts to 20 m.m. I could find these fossils in all places. Besides the stems I possess also a very fine *centrodorsalplate* of a calyx of *Millericrinus* originating from the clay-pit near Reuver, with the four sutures of fusion distinctly visible (the fused infrabasalia). Opposite to the round and roundish pentagonal, smooth or knobbed stems of the Apiocrines and Millericrines the remains of other *Crinoidea* are by far less numerous. From the boring 1 near Vlodrop I possess some free, starshaped segments of a species of *Pentacrinus*.

Of *Echinoidea* I have found only one spine in the clay-pit near Reuver. It is a typical spine of *Cidaris florigemma Phill.* of a length of 12 m.m. Some very thin and small plates of silex, covered with regularly arranged knobs, may also be derived from representatives of this group.

Very numerous again are all sorts of straight and curved small tubes of species of *Serpula* and *Terebella*, for which no further identification will be possible. Perhaps one spiral-rolled specimen from the boring 1 near Vlodrop can be considered as *Serpula convoluta Goldf.* while the rather common and faintly curved small tubes with a distinct parallel suture show much agreement with *Serpula limax Goldf.*

Of *Bryozoa* also some fragments are present: a nicely ramified little branch from the boring 1 and some other fragments consisting of small tubes placed radially from the Reuver and from a trialboring near Uden in North-Brabant. The silicification and the rounding however have rendered it impossible to identify those remains.

The *Brachiopoda* are represented by two specimina of a *Rhynchonella*: one very good specimen from the sandquarries of the Kollenberg near Sittard and a second incomplete specimen from a trial-boring at Reek near Grave. The Sittard specimen is closely allied to the *Rhynchonella 4-plicata Quenst.*, but shows on the ventral shell six radial ribs ending in the sinus of the frontal limb. It appeared impossible to me to identify this species with the literature I could dispose of [1]). The specimen of Reek does not allow any determination.

Little fragments of *Lamellibranchiata*-shells are not at all rare.

[1]) This specimen appeared later on to be identical with *Rhynchonella Thurmanni Voltz.* from the upper-Jurassic of French Lorraine.

The greater part belong to the *Ostreidae*; the smooth and flat fragments can be classed to the genus *Ostrea*, the severely folded and ribbed ones to *Alectryonia*. Among the latter *Alectryonia gregaria Sow.* may be represented. A fragment from Reuver suggests the delineation of *Trigonia costata Park.* Among the other fragments a radially ribbed printing reminds of *Cardium*, the rest however is totally irrecognisable.

Of *Gastropoda* I possess nothing else but the top of a real *Nerinea*-shell, from the clay-pit near Reuver, with distinct groove-line ("Schlitzband").

Finally of the *Cephalopoda* some fragments of *Belemnites* are at hand. The upper end of the rostrum of *Belemnites hastatus Blainv.* with alveole, found near Reuver, is rather well recognisable. The other fragments are very small and show no characteristic properties.

Summing up what is mentioned higher up we have the following list of fossils:

Miliolidae?	
Spongiae:	*Cnemidiastrum stellatum Goldf. sp.?*
	Eusiphonella Bronni Mst. sp.?
Anthozoa:	*Thamnastraea prolifera Becker.*
	Favia sp.?
Crinoidea:	*Apiocrinus*
	Millericrinus (f.i. M. horridus d'Orb.)
	Pentacrinus sp.?
Echinoidea:	*Cidaris florigemma Phill.*
Vermes:	*Serpula limax Goldf.?*
	Serpula convoluta Goldf.?
Bryozoa	
Brachiopoda:	*Rhynchonella Thurmanni Voltz.*
Lamellibranchiata:	*Ostrea sp.?*
	Alectryonia sp.?
	Alectryonia gregaria Sow.?
	Trigonia costata Park.?
	Cardium?
Gastropoda:	*Nerinea sp.?*
Cephalopoda:	*Belemnites hastatus Blainv.*

This fauna points without any doubt to an origin from younger *jurassic* strata, though few species can be identified with certainty. The fixing of the geological age by SCHLÜTER and FLIEGEL of the fossils from the places in the Lower Rhine basin (Duisdorf near Bonn, Weilerswist, browncoalmines Liblar and Donatus near Brühl etc.) appears to be also correct for those from the places in Limburg and North-Brabant, situated further to the North-West.

Mathematics. — "*Investigation of the functions which can be built up by means of infinitesimal iteration.*" By Mr. M. J. VAN UVEN. (Communicated by Prof. W. KAPTEYN).

(Communicated in the meeting of October 30, 1909).

In the paper presented by me in the preceding meeting I have roused the impression by an ambiguous mode of expression as if the method to obtain standardforms followed by me were the most serviceable. This is in nowise the case. I have exclusively intended to give a summary of what can be reached in this direction by differentiation. The last observation lying at hand and moreover already made by SCHROEDER according to which out of each function $y = \varphi(x)$ constructible by means of infinitesimal iteration a whole series of suchlike functions can be deduced all of the form $y = h_{-1}[\varphi\{h(x)\}]$ may have been made too much incidentally to weaken that impression properly. For, this last principle furnishes, as is immediately evident, an inexhaustible wealth of standardforms. With the aid of this principle we can i. a. just as well deduce the standardform (D). We have but to show that

$$y = \frac{\alpha x + \beta}{\gamma x + \delta}$$

follows directly out of the formula of ABEL or SCHROEDER. By applying the standardform of SCHROEDER:

$$g(y) = m\, g(x)$$

we find out of

$$y = y_{-1}\{m\, g(x)\} = \frac{\alpha x + \beta}{\gamma x + \delta},$$

$$g(x) = \frac{ax + b}{cx + d},$$

where m, a, b, c and d must satisfy

$$\frac{ad}{m\,\alpha - \delta} = \frac{bc}{\alpha - m\,\delta} = \frac{bd}{(1+m)\,\beta} = \frac{ac}{-(1+m)\,\gamma},$$

so that m is a root of

$$m^2 - \frac{\alpha^2 + \delta^2 + 2\,\beta\gamma}{\alpha\delta - \beta\gamma}\, m + 1 = 0.$$

We then find without difficulty the form (d) for $\varphi_n(x)$. In the parabolic case, $\alpha\delta - \beta\gamma = 0$, this calculation does not hold; we have then but to call in the aid of the equation of ABEL. Let us put

$$y = g_{-2}\{g(x) + 1\} = \frac{\alpha x + \beta}{\gamma x + \delta},$$

then follows out of it for $g(x)$

$$g(x) = \frac{ax + b}{cx + d},$$

where a, b, c and d must satisfy

$$\frac{ad - bc}{\alpha + \delta} = \frac{cd}{\alpha - \delta} = \frac{d^2}{2\beta} = \frac{c^2}{-2\gamma}.$$

In this case too the form (d') of $\varphi_n(x)$ can be determined without any difficulty.

The necessity of this supplement did not become quite clear to me until I received a communication of Dr. L. E. J. BROUWER, who was so kind as to draw my attention to this lack and who likewise brought out the immediate connection between the standardform (D) and that of SCHROEDER. I set great store by the fact of thanking him kindly for his observations.

Physics. — "*Remarks on the Experiments of* WILSON *and* MARTYN *on the Velocity of Rotation of the Electric Discharge in Gases in a Radial Magnetic Field.*" By Dr. J. A. VOLLGRAFF. (Communicated by Prof H. A. LORENTZ).

(Communicated in the meeting of October 30, 1909).

WILSON and MARTYN [1]) have proposed a theory on the rotation of the electric discharge in gases at low pressures in a radial magnetic field, agreeing satisfactorily with their experiments. The discharge consists in a motion of positive and negative ions.

They find for the value of the velocity of rotation

$$U = k_1 k_2 H X, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

H being the magnetic and X the electric intensity at the place of the discharge, and k_1 and k_2 the velocities which unit electric intensity acting on the gas gives to a positive and a negative ion resp.

The apparatus was composed of two vertical coäxial cylinders of glass C_1 and C_2 along the axis of which an electromagnet was so placed that one of its poles was situated in the middle of the magnet and at the same time in the middle of the axis. The magnetic field

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[1]) H. A. WILSON and G. H. MARTYN. "On the Velocity of Rotation of the Electric Discharge in Gases at Low Pressures in a Radial Magnetic Field". (Proc. Roy. Soc. Ser. A. Vol. 79, N° A 532, 2 Aug. 1907).

in every plane cutting the axis at right angles was nearly radial, the magnetic intensity therefore, on account of its solenoidal distribution, being inversely proportional to the distance from the axis. The basis and the upper surface of the two cylinders consisted in two horizontal metal plates. The discharge passed nearly vertically from a point of the upper plate to a point of the basis, and was carried round about the axis describing a third coaxial cylinder.

If the space between the two cylinders C_1 and C_2 had been filled up with an electrolyte, the whole mass would have rotated under the influence of the magnetic field; the same being true of course if the motion of the charged particles had taken place in a solid body.

In the experiments of WILSON and MARTYN the gas must likewise have been carried along, but the motion of the gas must have been so slow that the discharge may indeed be considered to have revolved about the axis in a medium at rest. The truth of this assertion appears if we calculate the velocity of the gas and compare it with the observed velocity of the discharge.

As to the formula (1), we can show that it may be obtained in another way than WILSON and MARTYN's. The mechanism of the discharge is not necessarily such as they assume it to be.

§ 1. Let the radius of the larger cylinder be R_1 that of the smaller one R_2. Let us suppose that the discharge does not start from one point only but simultaneously from all points situated between two cylinders, coaxial with the former, of radius ϱ_1 and ϱ_2 resp. ($\varrho_1 > \varrho_2$). A circular discharge of this kind was indeed observed in some of the experiments. It is a reasonable supposition that when the discharge does start from one point only, the velocity of revolution of the gas is of the same order of magnitude as the velocity here calculated.

Let I be the total current. Since the section of the discharge is $\pi(\varrho_1^2 - \varrho_2^2)$, the density of the current is

$$\frac{I}{\pi(\varrho_1^2 - \varrho_2^2)} \quad \ldots\ldots\ldots\ldots \quad (2)$$

It follows that the current between the cylinders with radius r and $r + dr$ resp. is

$$di = \frac{2r\,dr}{\varrho_1^2 - \varrho_2^2} I \quad \ldots\ldots\ldots\ldots \quad (3)$$

if $\varrho_1 > r >$.

Let ω be the angular velocity of the gas when the stationary state is reached.

If two cylinders with radius r and $r + dr$ resp. are so situated that $r > \varrho_1$ and $\varrho_2 > r + dr$, whence it follows that there is no current through the space which they include, the moments of the friction in the gas on the surfaces of these two cylinders will be equal and opposite in the stationary state.

If $v = \omega r$ is the velocity, and η the coëfficient of friction, these moments per unit of length measured in a direction parallel to the axis, are resp.

$$-2\pi r^2 . \eta \frac{dv}{dr} \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

and

$$+ 2\pi (r + dr)^2 \eta \left(\frac{dv}{dr}\right)_{r+dr} . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

Since they are in equilibrium

$$-r^2 \frac{dv}{dr} + (r + dr)^2 \left(\frac{dv}{dr}\right)_{r+dr} = 0, \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

or

$$2\omega + 4r \frac{d\omega}{dr} + r^2 \frac{d^2\omega}{dr^2} = 0 . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

The solution of this equation is

$$\omega = \frac{a}{r} + \frac{b}{r^2} . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

The friction at the cylindrical surfaces bounding the gas is assumed to be so great that the angular velocity there is zero.

We have therefore outside the space occupied by the discharge

$$\omega = b_1 \left(\frac{1}{r^2} - \frac{1}{rR_1}\right) . \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

and inside that space

$$\omega = b_2 \left(\frac{1}{r^2} - \frac{1}{rR_2}\right), \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

b_1 and b_2 being constants.

If however the radii r and $r + dr$ are smaller than ϱ_1 and greater than ϱ_2, the moment $Hrdi$ acts on the gas between the cylinders with radii r and $r + dr$, on account of the current flowing there.

This moment can be written in the form

$$\frac{2Hr^2\, dr}{\varrho_1^2 - \varrho_2^2} I . \quad . \quad . \quad . \quad . \quad . \quad . \quad (11)$$

Considering the moments of the friction, we therefore obtain

$$\frac{-2HI\,r^2dr}{\varrho_1^{\,2}-\varrho_2^{\,2}} = 2\pi\,.\,\eta\left[-r^2\,\frac{dv}{dr}+(r+dr)^2\left(\frac{dv}{dr}\right)_{r+dr}\right] \quad . \quad . \quad (12)$$

or

$$\frac{-2HIr^2dr}{\varrho_1^{\,2}-\varrho_2^{\,2}} = 2\pi\,.\,\eta r dr\left[2\omega+4r\,\frac{d\omega}{dr}+r^2\,\frac{d^2\omega}{dr^2}\right] \quad . \quad . \quad . \quad (13)$$

So we have

$$\frac{-HIr}{\pi\eta(\varrho_1^{\,2}-\varrho_2^{\,2})} = 2\omega+4r\,\frac{d\omega}{dr}+r^2\,\frac{d^2\omega}{dr^2} \quad . \quad . \quad . \quad . \quad (14)$$

If we put

$$\frac{HIr}{2\pi\eta(\varrho_1^{\,2}-\varrho_2^{\,2})} = C, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (15)$$

which is a constant, since H is inversely proportional to r, the solution of equation (14) is

$$\omega = \frac{a}{r}+\frac{b}{r^2}-C. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (16)$$

Since the angular velocity must be continuous for $r=\varrho_1$ and for $r=\varrho_2$ we obtain from the formulae (9), (10) and (16)

$$\frac{a}{\varrho_1}+\frac{b}{\varrho_1^{\,2}}-b_1\left\{\frac{1}{\varrho_1^{\,2}}-\frac{1}{\varrho_1 R_1}\right\} = C, \quad . \quad . \quad . \quad . \quad (17)$$

and

$$\frac{a}{\varrho_2}+\frac{b}{\varrho_2^{\,2}}-b_2\left\{\frac{1}{\varrho_2^{\,2}}-\frac{1}{\varrho_2 R_2}\right\} = C. \quad . \quad . \quad . \quad . \quad (18)$$

Besides $\frac{d\omega}{dr}$ must be continuous. This can be proved by considering a flat cylindrical space containing a part of the surface bounding the space occupied by the discharge. The flat space is so chosen that its two parallel and equal surfaces are also parallel to the said surface and are situated at an infinitely small distance from that surface on either side of it. The force exerted by the magnetic field on the ions in the flat space may be neglected, so there must be equilibrium between the forces of friction on its two parallel surfaces, whence we infer that there must be continuity of $\frac{dv}{dr}$ and since ω is continuous, of $\frac{d\omega}{dr}$.

Hence

$$\frac{a}{\varrho_1}+\frac{2b}{\varrho_1^{\,2}}-b_1\left\{\frac{2}{\varrho_1^{\,2}}-\frac{1}{\varrho_1 R_1}\right\} = 0 \quad . \quad . \quad . \quad . \quad . \quad (19)$$

and

$$\frac{a}{\varrho_2} + \frac{2b}{\varrho_2^2} - b_2\left\{\frac{2}{\varrho_2^2} - \frac{1}{\varrho_2 R_2}\right\} = 0 \quad . \quad . \quad . \quad . \quad . \quad (20)$$

From (17) and (19) we obtain

$$\frac{b}{\varrho_1^2} - \frac{b_1}{\varrho_1^2} + C = 0, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (21)$$

from (18) and (20)

$$\frac{b}{\varrho_2^2} - \frac{b_2}{\varrho_2^2} + C = 0. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (22)$$

So

$$b = b_1 - C\varrho_1^2 = b_2 - C\varrho_2^2 \quad . \quad . \quad . \quad . \quad . \quad . \quad (23)$$

The equation (17) becomes

$$a = -\frac{b}{R_1} - C\varrho_1\left(\frac{\varrho_1}{R_1} - 2\right) \quad . \quad . \quad . \quad . \quad . \quad . \quad (24)$$

and the equation (18) becomes

$$a = -\frac{b}{R_2} - C\varrho_2\left(\frac{\varrho_2}{R_2} - 2\right). \quad . \quad . \quad . \quad . \quad . \quad (25)$$

From (24) and (25) we obtain

$$b = C\,\frac{R_1\varrho_2\,(2R_2 - \varrho_2) - R_2\varrho_1\,(2R_1 - \varrho_1)}{R_1 - R_2} \quad . \quad . \quad . \quad . \quad (26)$$

and

$$a = C\,\frac{\varrho_1\,(2R_1 - \varrho_1) - \varrho_2\,(2R_2 - \varrho_2)}{R_1 - R_2} \quad . \quad . \quad . \quad . \quad . \quad (27)$$

Substituting these values in (16) we find

$$\omega = C\left\{\frac{\varrho_1(2R_1 - \varrho_1) - \varrho_2(2R_2 - \varrho_2)}{(R_1 - R_2)r} + \right.$$

$$\left. + \frac{\varrho_2 R_1(2R_2 - \varrho_2) - \varrho_1 R_2(2R_1 - \varrho_1)}{(R_1 - R_2)r^2} - 1\right\} \quad . \quad . \quad . \quad (28)$$

In the peculiar case in which ϱ_1 and ϱ_2 differ very little from one another we may put

$$\left.\begin{aligned} \varrho_2 &= \varrho \\ \varrho_1 &= \varrho + \delta \\ r &= \varrho + \varepsilon \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad . \quad . \quad (29)$$

δ and ε being infinitely small. In this case we have

$$C = \frac{HI}{4\pi\eta\,.\,\delta} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (30)$$

and

$$\omega = 2C\sigma \frac{(\varrho - R_2)(R_1 - \varrho)}{\varrho^2 (R_1 - R_2)} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (31)$$

(ε drops out).

Substituting (30) in (31) we find

$$\omega = \frac{HI}{2\pi\eta} \cdot \frac{(\varrho - R_2)(R_1 - \varrho)}{\varrho^2 (R_1 - R_2)} \quad . \quad . \quad . \quad . \quad . \quad . \quad (32)$$

Since the discharge in the experiments under consideration had the form of a small column, and since the values of the field H are mentioned for one peculiar value of r (viz. $r = 1.34$ cm.) we may use the formula (32) putting $\varrho_1 = \varrho_2 = \varrho = 1.34$. In the apparatus of WILSON and MARTYN R_1 was equal to 1.505 cm. and R_2 to 1.16 cm.

The value of η is independent of the pressure. Say for nitrogen $\eta = 0.0002$. In one of the experiments made with nitrogen the value of I was between 0.001 and 0.003 (el. magn. unities) and the intensity of the magnetic field was 41.

Substituting these values in (32), we find that the angular velocity with which the gas moved in that experiment, supposing it to be the same as if the discharge had the form of a complete cylinder, must have been between 1.6 and 4.7. As to the observed angular velocity of the discharge, it was $2\pi \,.\, 9.5$ or about 60.

In the same way we find by substituting numerical values belonging to other experiments that the angular velocity of the gas was always much inferior to that of the discharge itself.

The general expression of the angular velocity of the discharge for $r = \varrho$ is after (1)

$$\Omega = \frac{k_1 k_2}{\varrho} H X, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (33)$$

So the proportion of the two angular velocities is found to be

$$\frac{\Omega}{\omega} = 2\pi\eta k_1 k_2 \frac{\varrho (R_1 - R_2)}{(\varrho - R_2)(R_1 - \varrho)} \cdot \frac{X}{I}, \quad . \quad . \quad . \quad . \quad (34)$$

or, since $I = \sigma \,.\, \pi (\varrho_1^2 - \varrho_2^2) X$, σ being the specific conductivity,

$$\frac{\Omega}{\omega} = 2 \frac{\eta}{\sigma} k_1 k_2 \frac{\varrho (R_1 - R_2)}{(\varrho_1^2 - \varrho_2^2)(\varrho - R_2)(R_1 - \varrho)} \quad . \quad . \quad . \quad . \quad (35)$$

This proportion increases as $\varrho_1 - \varrho_2$ diminishes.

§ 2. The same formulae (28) and (33) are applicable to the case of an electrolyte, instead of a gas, filling the space between the cylinders C_1 and C_2.

In that case the discharge will not be limited to a thin hollow cylinder, but will fill the whole space between the cylinders C_1 and

C_2. We must therefore put $\varrho_1 = R_1$ and $\varrho_2 = R_2$ in (28). Hence

$$\omega = \frac{HI}{2\pi\eta} \cdot \frac{(r-R_2)(R_1-r)}{r(R_1^2-R_2^2)} \quad . \quad . \quad . \quad . \quad . \quad . \quad (36)$$

The maximum-value of ω is reached for $r = \frac{2R_1R_2}{R_1+R_2}$, Hr still being a constant.

This maximum-value is therefore

$$\omega_M = \frac{HI}{4\pi\eta} \cdot \frac{(R_1-R_2)}{(R_1+R_2)^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad (37)$$

and the proportion of the angular velocity of the ions carrying the current to that of the whole electrolyte, both taken for $r = \frac{2R_1R_2}{R_1+R_2}$, is found to be

$$\frac{\Omega}{\omega_M} = 2\pi\eta_l k_1 k_2 \frac{(R_1+R_2)^3}{R_1R_2(R_1-R_2)} \cdot \frac{X}{I}, \quad . \quad . \quad . \quad . \quad (38)$$

or, since $I = \sigma \cdot \pi(R_1^2 - R_2^2)X$,

$$\frac{\Omega}{\omega_M} = 2\eta_l k_1 k_2 \frac{1}{\sigma} \frac{(R_1+R_2)^2}{R_1R_2(R_1-R_2)^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad (39)$$

If e is the charge of a gram-ion and if each cm^3 contains n gram-ions, we find

$$\sigma = en(k_1 + k_2), \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (40)$$

so that (39) becomes

$$\frac{\Omega}{\omega_M} = 2\frac{\eta_l}{en} \frac{k_1k_2}{k_2+k_2} \frac{(R_1+R_2)^2}{R_1R_2(R_1-R_2)^2} \quad . \quad . \quad . \quad . \quad . \quad (41)$$

Put $R_1 = 1{,}505$ cm. and $R_2 = 1.16$ cm. Take

$$\eta_l = 0{,}01$$

(coefficient of friction of water).

We have

$$e = 9654.$$

If the electrolyte consists in a solution of 10 Gr. of KCl in a Litre,

$$n = \frac{1}{7460}.$$

The velocities for K and Cl are $k_1 = 0{,}66.10^{-11}$ and $k_2 = 0{,}69.10^{-11}$ resp. Substituting these values in the equation (41) we find a very small value for $\frac{\Omega}{\omega_M}$ (about 7.10^{-13}).

Since the angular velocity is so small in this case, the electrolyte will acquire nearly the same velocity as the ions carrying the current. Unlike what happens in the case of a gas, the ions carrying the

current have a velocity superior by a small value only to that of the medium through which they move.

§ 3. WILSON and MARTYN calculate the velocity of the rotating discharge very nearly in the following manner.

If X is the electric intensity parallel to the axis of the cylinder, positive and negative ions will have the velocities $k_1 X$ and $-k_2 X$ in the direction of that intensity. Say that the positive ions move downward with the velocity

$$v_1 = k_1 X, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (42),$$

and the negative ones upward with the velocity

$$v_2 = k_2 X. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (43)$$

The mechanical force acting on each ion is equal to the product of the magnetic intensity, the charge of the ion and its velocity. Thence we conclude that the velocities with which the positive and negative ions resp. move laterally, are

$$u_1 = k_1^2 HX. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (44)$$

and

$$u_2 = k_2^2 HX. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (45)$$

Positive and negative ions will be able to follow the same path only when it is inclined, the part near the anode being in advance. Let α be the angle which the discharge makes with the axis. This angle will appear to be very small.

If V_1 is the velocity of a positive ion in the direction of the discharge and U the lateral velocity of the discharge, the true velocity of the positive ion will be at the same time the resultant of the velocities v_1 and u_1 and of the velocities V_1 and U.

We must have therefore

$$v_1 = V_1 \cos \alpha \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (46)$$

and

$$u_1 = U - V_1 \sin \alpha \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (47)$$

And if the negative ions move upward in the direction of the discharge with the velocity V_2 we must likewise have

$$v_2 = V_2 \cos \alpha \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (48)$$

and

$$u_2 = U + V_2 \sin \alpha \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (49)$$

From the four last equations we obtain

$$tg\, \alpha = \frac{u - u_1}{v_1} = \frac{u_2 - U}{v_2}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (50)$$

$$U = \frac{v_1 u_2 + v_2 u_1}{v_1 + v_2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (51)$$

and

$$tg\,\alpha = \frac{u_2 - u_1}{v_1 + v_2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (52)$$

Substituting the values of v_1, v_2, u_1, and u_2 according to (42), (43), (44) and (45) in (51) and (52), we find

$$U = k_1 k_2 HX \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (53)$$

and

$$tg\,\alpha = (k_2 - k_1)H \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (54)$$

Considering the values of k_1 and k_2 to be known, and being able to measure U and X (X roughly indeed), we may verify the formula (53). But if k_1 and k_2 are considered as unknown quantities the formula may serve to find the product $k_1 k_2$.

From other experiments the values of k_1 and k_2 are sufficiently well known. Substituting these values in the formula (54) we find that the angle α differs very little from zero, so that the inclination will not be visible.

Now one can ask whether positive and negative charges will not be formed in the front and at the back of the discharge, as in HALL's phenomenon.

We suppose until further notice that the path of the discharge is parallel to the axis of the cylinders and that the said charges have been formed, in consequence of which the ions are acted upon by a horizontal electric intensity Y, X being again the vertical electric intensity. The velocities v_1 and v_2 will be as great as in the former case, but the horizontal velocities of the positive and negative ions become resp.

$$u_1 = k_1^2 HX + k_1 Y \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (55)$$

and

$$u_2 = k_2^2 HX - k_2 Y \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (56)$$

Both of them must now be equal to U, the velocity of the discharge. So we find

$$U = k_1^2 HX + k_1 Y = k_2^2 HX - k_2 Y \quad . \quad . \quad . \quad . \quad . \quad (57)$$

whence

$$Y = (k_2 - k_1) HX \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (58)$$

and

$$U = k_1 k_2 HX \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (59)$$

The same value as before is found in this way for the velocity U. It is quite possible that the path of the discharge has an *arbitrary*

small inclination β and that electric charges exist at the front and at the back of the discharge.

In that case we again find the formula (51), in which v_1 v_2 u_1 and u_2 are given by the formulae

$$\left.\begin{aligned} v_1 &= k_1 X \\ v_2 &= k_2 X \\ u_1 &= k_1{}^2 HX + k_1 Y \\ u_2 &= k_2{}^2 HX - k_2 Y \end{aligned}\right\} .$$

The place of (52) is taken by the formula

$$tg\,\beta = \frac{u_2 - u_1}{v_1 + v_2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (60)$$

From (51) and (60) we conclude

$$U = k_1 k_2 HX \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (61)$$

and

$$tg\,\beta = (k_2 - k_1) H - \frac{Y}{X} \quad . \quad . \quad . \quad . \quad . \quad . \quad (62)$$

Whatever the inclination may be, we always find the right value for the velocity U. So it appears reasonable to suppose that this inclination is determined by the local conditions of the metal and that the electric intensity Y and the charges to which it is due, adjust themselves to it according to (62).

Physics. — *"The atomic volume of allotropic modifications at very low temperatures."* By Prof. Ernst Cohen and Dr. J. Olie Jr. Communication N°. 113 from the Physical Laboratory at Leiden. (Communicated by Prof. H. Kamerlingh Onnes).

(Communicated in the Meeting of October 30, 1909).

1. In the discussion on a communication: "The allotropic forms of silver and gold", made by one of us in the meeting of the Deutsche Bunsengesellschaft at Dresden [1]) the question was raised whether the atomic volumes of the different allotropic modifications of a certain element become equal at the absolute zero point. As the answer to this question, which can only be given by experiment, is in the closest connection with some other points concerning the periodic system of the elements to which we shall come back later on, we have carried out an investigation in the above mentioned direction, which may be briefly described here.[2])

[1]) Zeitschrift für Elektrochemie **21**, 589 (1906).

[2]) The full discussion will shortly appear in the Zeitschrift für physik. Chemie

2. We have carried out our measurements only with diamond and graphite, and with white and gray tin, as a preliminary investigation showed us, that the allotropic forms of phosphorus were less suitable.

3. Through the kindness of Mr. S. LEHMANS of Amsterdam Mr. LOUIS TAS placed ten grammes of diamond (314 pieces) at our disposal. We may be allowed here also to express our hearty thanks to them.

4. Mr. P. LEBEAU of Paris sent us a large quantity of graphite, made by the late HENRI MOISSAN; on investigation this preparation appeared to be very pure.

As the researches of LE CHATELIER and WOLOGDINE [1]) have shown that graphite assumes a sharply defined specific gravity at a certain temperature only when it has been exposed to great pressures (according to the above writers enclosed gases are expelled from it in this way), we have exposed our preparation to pressures of 5000—1000 atms., and have not used it until the spec. grav. remained constant after repeated compression.

We made use of an apparatus constructed specially for this purpose, which is reproduced in fig. 1.

In the steel block B a hole is bored, into which powdered graphite is put. In this hole fits the steel piece D, which is pressed down through the block A under an hydraulic press. The graphite cylinder made in this way may be easily removed from B, if C is unscrewed, and may then be pushed from the hole by means of the piece FE.

5. The white tin, which was used in our measurements, was the same preparation that served in the investigations of ERNST COHEN and E. GOLDSCHMIDT [2]); the grey tin came from a block of Banka tin, which on a former occasion had been kindly presented to us by Mr. H. BAUCKE, chemical engineer [3]). On investigation this material proved to be very pure. By a special investigation we convinced ourselves that it did not contain any white tin.

DETERMINATION OF THE SPECIFIC GRAVITY.

6. Our determinations were carried out at 18°, —38°, and —164°.

A. *Measurements at* 18°.0.

7. For this purpose we made use of a pycnometer. As graphite is not moistened by water, we took toluene as filling liquid. In

[1]) C. R. **146**, 49 (1908).
[2]) Zeitschrift fur physik. Chem. **50**, 225 (1905).
[3]) Zeitschrift für physik. Chem. **63**, 625 (1908).

connection with this the ground-glass stopper of the pycnometer was made very long. Our apparatus had a capacity of ± 22 cc.; it was gauged by means of water carefully freed from gases by boiling. The weighings were carried out on a balance, on which $\frac{1}{10}$ mg. could be read. All weighings were reduced to vacuum and to the density of water at 4°.0.

After the substance which was to be examined, had been introduced into the pycnometer, the latter was filled with toluene, and connected with the air pump. When the toluene had boiled for some time, the pycnometer was again filled up with thatliquid, and placed in a thermostat, which was kept at 18°.0. The thermometers used were tested by a normal thermometer (divided into $^1/_{10}^\circ$), which had been tested by the Physikalisch-Technische Reichsanstalt at Charlottenburg.

The stopper of the pycnometer had a capillary bore; the upper surface of the capillary was made opaque by grinding. As soon as this upper surface remained perfectly dry for ten minutes, the liquid was sucked off by means of a capillary pipette down to a mark on the capillary of the pycnometer. Then the pycnometer was removed from the thermostat, and weighed after having been carefully dried. All the determinations were made twice.

For the toluene used we found: $d\,\frac{18°.0}{4°.0}$ **0,8666**.

Table I contains the results of the determinations at 18°.0.

TABLE 1[1])

Temperature 18°.0.

Substance		Weight of the expelled toluene in gr.	Weight of the expelled water in gr.	d. $\frac{18°.0}{4°.0}$	Remarks
Name	Weight in gr.				
Diamond	9.9790*	—	2.8296*	3.514	
Graphite	8.4424*	3.2964*	—	2.217	
Graphite	8.5504*	3.3412*	—	2.215	
White tin	16.8488	—	2.3082	7.281	prepar. from grey tin
White tin	23.7052*	—	3.2458*	7.285	„ from melted tin
Grey tin	33.4100*	—	5.7953*	5.751	fine powder
Grey tin	29.9170	—	5.1789	5.763	coarser „

[1]) The values in this table marked with an * have served in the further calculations.

B. *Measurements at low temperatures.*

8. As far as we know an accurate method of determining the specific gravities of solid substances at very low temperatures does not exist yet [1]), we therefore had in the first place to devise one.

Our method may be considered as a combination of the dilatometer and the weight-thermometer.

It appeared at the outset that we could extend our experiments no further than $-164°$. For no liquid is known as yet which remains sufficiently fluid below that temperature. Pentane e. g. (so-called Pentan für Thermometer) becomes so viscous a few degrees lower, that it is of no use whatever.

a. *The dilatometer.*

9. Our dilatometer (Fig. 2) was made of Jena glass 16^{III}, as the coefficient of expansion of this kind of glass at very low temperature is sufficiently known through the investigations of KAMERLINGH ONNES and CLAY.

A is a reservoir of a capacity of $\pm$ 20 cc.; the capillary BB_1B_2 has a bore of 0,7 mm.; the capacity of C is $\pm$ 6 cc., that of E $\pm$ 20 cc.

By the aid of a ground-glass junction S, E could be placed on the capillary tube. This has the same bore as BB_1B_2.

The course of the experiment is now as follows:

After A has been cleaned and weighed, the substance which is to be examined as to its specific gravity, is introduced, and the whole is weighed again. The tube BB_1B_2 is fused on to it and the whole weighed again. Then a mark is etched on the capillary BB_1B_2, just below the place where the level of the cooling liquid will ultimately be. A is filled with pentane up to this mark, after it has been brought to 18°.0 in a thermostat. It is weighed again.

Now A is put into a refrigerating mixture, and the other part of the apparatus is fused to the capillary at B_2.

E is connected with a three-way tap (*not* greased), which enables us to exhaust the apparatus, and to fill it up with pentane. These manipulations are repeated a few times.

If in this way the apparatus has been filled with pentane to about halfway the height of E, we pour pure, doubly distilled mercury

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[1]) The determinations made by DEWAR, Chemical News **85**, 289 (1902), were of a more preliminary character. His method was altogether unsuitable for our purpose as it is not sufficiently accurate.

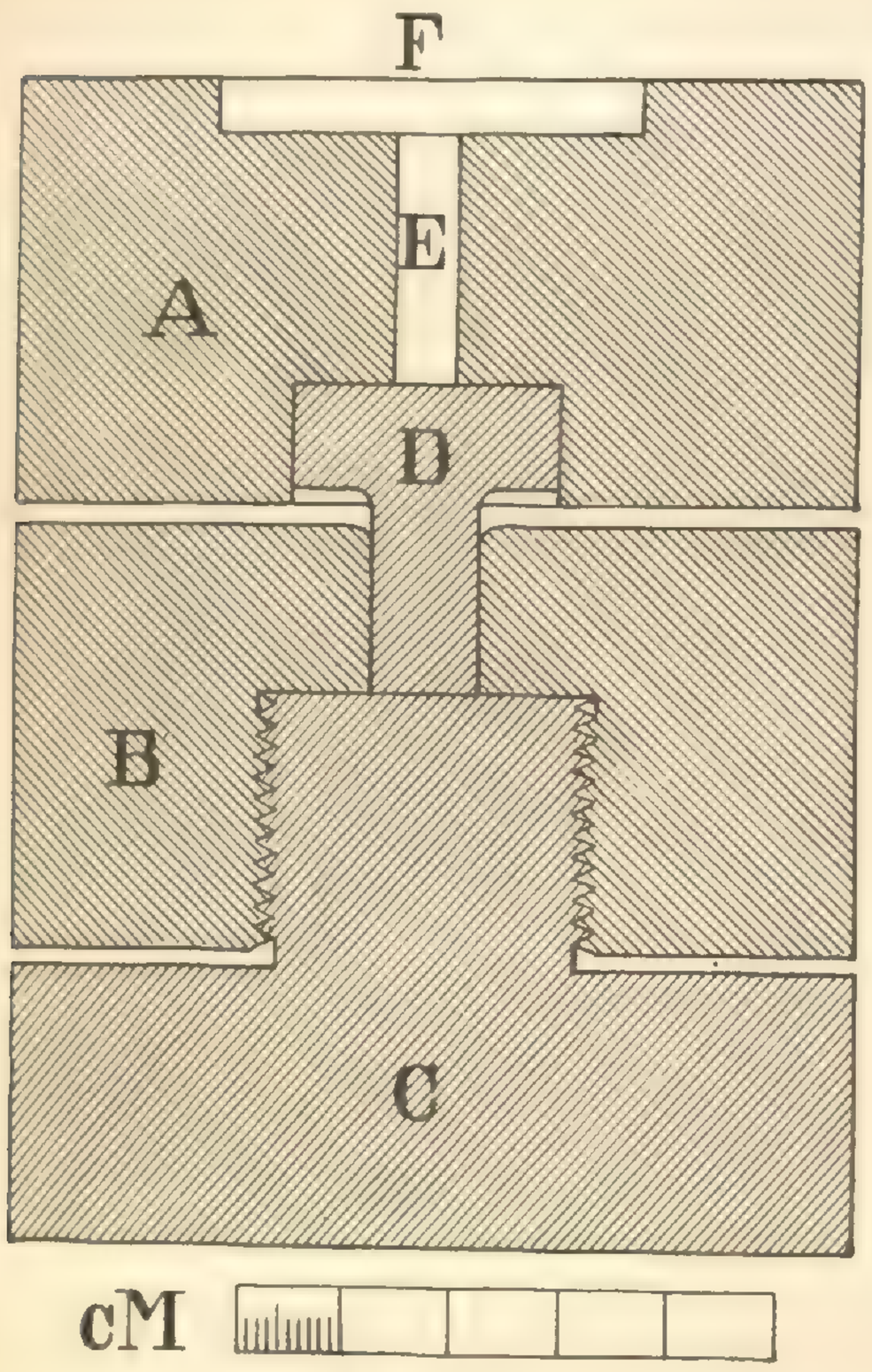

Fig. 1.

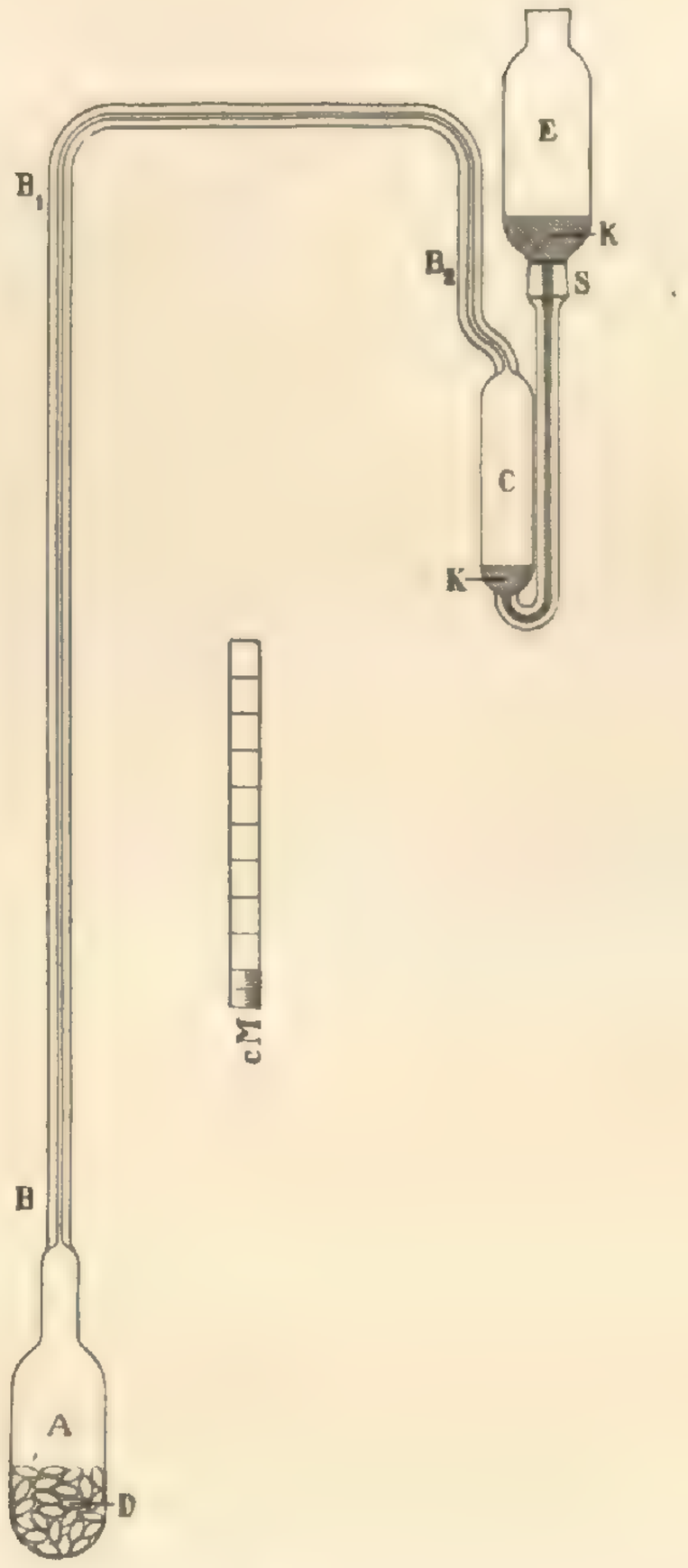

Fig. 2.

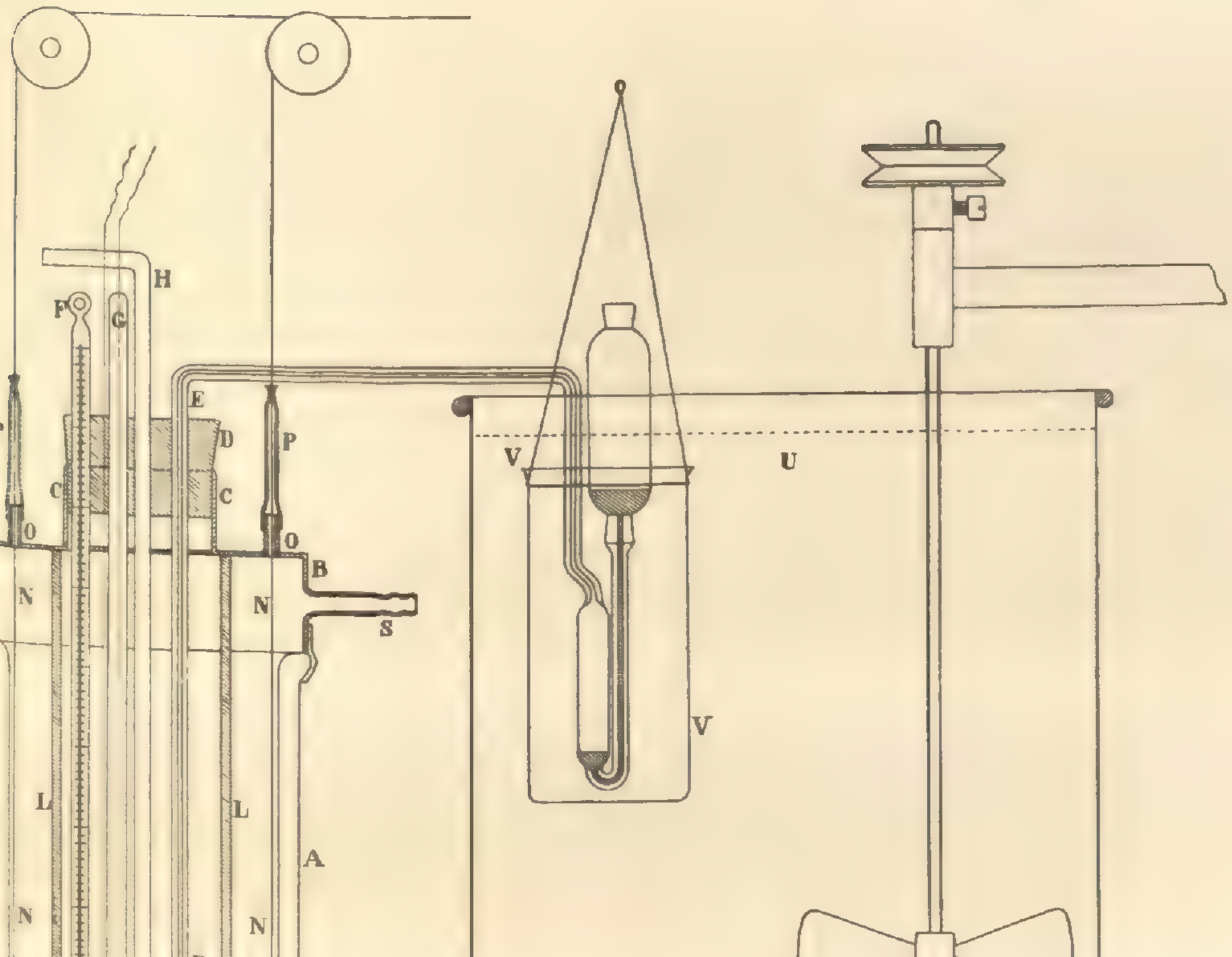

Fig. 3.

into E by means of a funnel. We remove the excess of pentane by exhausting once more, so that the mercury at last penetrates into the bottom part of C, and KS remains filled with it.

ABB_1 is placed in the cryostat, and B_2CSE in a thermostat U (fig. 3), which is kept at 18°.0.

The mercury now rises in C; mercury is poured into E, so that, when A has assumed the temperature of the cryostat, the mercury in E stands a few centimeters above the ground-glass junction.

If the temperature of the cryostat has remained perfectly constant for an hour (the temperature is determined by means of a resistance thermometer), E is removed. The excess of mercury in E, is then received in a glass beaker V (fig. 3). If the capillary remains quite filled to the top, the vessel E is again placed on the ground-glass junction after having been care fully cleaned beforehand, and now the temperature of ABB_1 is slowly raised. The mercury that issues at S, is received in E. After some time the whole apparatus is placed in the thermostat of 18°.0, and there it is left for a considerable time. Now a glass beaker is placed under B_1CSE, E is carefully removed from the ground-glass junction, so that the mercury flows into the beaker, the mercury on the surface of the ground junction being collected with a feather.

The mercury found in the beaker is carefully washed with water, then with alcohol. This is removed by means of a current of air which has been filtered through cotton wool. The drying is repeated, till the weight has become constant.

b. *The Cryostat.*

10. For the measurements at $-38°$ we used liquid methyl chloride under reduced pressure, at $-164°$ liquid methane at 1 atm. The apparatus constructed by KAMERLINGH ONNES for such purposes [1]), is diagrammatically represented in fig. 3. AA is an entirely transparent vacuum glass, which can contain $\pm$ 2 liters of liquid, BB is a brass cover, which can be fastened gastight on the glass by means of a rubber ring. The neck CC can be closed gastight by means of a large rubber stopper. The stopper D was pierced in six places; through three holes passed the capillaries E of three dilatometers. Moreover the glass vessel AA was provided with a resistance thermometer G, a pentane thermometer F, and a glass tube H. The last reached to the bottom of the vacuum glass.

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[1]) These Proc. June 1905; Comm. from the Phys. Lab. at Leyden N°. 94*c*.

The reservoirs T of the dilatometers were surrounded by a protecting jacket $KKKK$ of nickel plate. This jacket was supported by the nickel bars LL. The liquid gas was kept in violent motion by the stirrer mm. The valves rrr promote the stirring in a high degree. The stirrer is suspended on three wires NN, which pass airtight through the cover BB. To satisfy this condition they pass through the metal tubes OO, and through the rubber tubes PP; in the latter the wires are fastened airtight. By means of an electromotor the wires are brought in an upward and downward motion.

The liquid gas was pumped into the vacuum glass by means of the tube HH.

When we worked under reduced pressure, S was connected with the airpump.

11. The tables 2—9 contain the results of the measurements, which are used in the calculation.

TABLE 2.

Temperature 18.°0.

The dilatometer contains	Weight of the solid substance in gr.	Weight of the pentane in gr.	Number of the dilatometer
Diamond—Pentane	9.9790	12.0820	I
Graphite—Pentane	8.7954	11.6135	II
White tin—Pentane	23.7026	12.7496	III
Grey tin—Pentane	26.4843	10.5339	IV
Pentane	—	12.8847	V
Water	—	20.2058	V

TABLE 3.

Temperature —163°.6.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0.
I	Diamond– Pentane	67.5817
II	Graphite—Pentane	65.0153
V	Pentane	72.1680

TABLE 4.

Temperature —163°.2.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0
I	Diamond—Pentane	67.6155
II	Graphite—Pentane	65.0644
V	Pentane	72.2079

TABLE 5.

Temperature —163°.5.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0
I	Diamond - Pentane	67.5664
II	Graphite—Pentane	65.0778
V	Pentane	72.2685

TABLE 6.

Temperature —38°.0.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0
I	Diamond—Pentane	21.2138
II	Graphite—Pentane	20.4593
V	Pentane	22.7002

TABLE 7.

Temperature —37°.75.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0
I	Diamond—Pentane	21.1658
II	Graphite—Pentane	20.3958
V	Pentane	22.6509

TABLE 8.

Temperature —163°.3.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0
III	White tin—Pentane	71.7180
IV	Grey tin—Pentane	58.9882
V	Pentane	72.2378

TABLE 9.

Temperature — 163°.4.

Number of the dilatometer	The dilatometer contains	Weight in gr. of the mercury that has flowed out at 18°.0
III	White tin—Pentane	71.7131
IV	Grey tin—Pentane	58.9744
V	Pentane	72.2318

12. The calculation of the experiments may be summarized here as follows: if we call:

P_d the weight of the diamond used;

$P^{Ip}_{18°.0}$ the weight of the pentane at 18°.0 in the dilatometer I with the diamond;

$P^{V}_{18°.0}$ the weight of the pentane, in the dilatometer V at 18°.0:

U_p the weight of the mercury which has flowed out during the heating of dilatometer V (filled with pentane) from $-t°$ to 18°.0:

$S_{18°.0}$ the specific gravity of mercury at 18°.0;

$W^{d}_{18°.0}$ the weight of the water that is expelled by the diamond at 18.°0.

$D^{w}_{18°.0}$ the density of water at 18.°0;

δ the mean coefficient of expansion of the (Jena) glass used from 18° to $-t°$.

U_d the weight of the mercury which has flowed from dilatometer I, when it is heated from $-t°$ to 18°.0, then the specific gravity of diamond at $-t°$ is

$$S_{-t°} = \frac{P_d}{\frac{P^{Ip}_{18°.0}}{P^{V}_{18°.0}}\frac{U_p}{S_{18°.0}} + \frac{W^{d}_{18°.0}}{D^{w}_{18°.0}}(1-\delta(t+18°)) - \frac{U_d}{S_{18°.0}}}$$

Table 10 has been calculated by means of this expression (resp. by means of the analogous expressions for the other substances investigated.

TABLE 10.

Temperature t	Spec. gravity $d\frac{t}{4°.0}$			
	Diamond	Graphite	White tin	Grey tin
— 163°.6	3.519	2.223		
— 163°.2	3.518	2.224		
— 163°.5	3.509	2.222		
— 163°.3			7.350	5.768
— 163°.3			7.351	5.768
— 38°.0	3.510	2.217		
— 37°.75	3.510	2.217		
+ 18°.0	3.514	2.216	7.285	5.751

13. Finally table 11 gives a survey of the ratio of the spec. gravities of different allotropic forms of the same element at different temperatures:

TABLE 11.

Temperature	Ratio $\frac{S_{diamond}}{S_{graphite}}$	Ratio $\frac{S_{white\ tin}}{S_{grey\ tin}}$
+ 18°.0	1.585	1.266
— 38°	1.583	
— 164°	1.582	1.274

14. We see from table 11 that down to — 164° there are no indications that the spec. gravities (resp. the spec. volumes) of the different allotropic modifications of the same element converge to one and the same value in approaching the absolute zero point.

It is only owing to the great kindness of Prof. KAMERLINGH ONNES, who placed the resources of his laboratory at our disposal, that it has been possible for us to carry out the above investigation. We may be allowed also here to express our great indebtedness to him.

To Mr. G. J. FLIM, technical assistant at the Cryogenic Laboratory at Leiden, many thanks are due for his intelligent assistance.

October 1909.

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Physics. — *"On the origin of the chromospheric light."* By Prof. W. H. JULIUS.

In a paper, entitled: Photography of the "flash" spectrum without an eclipse [1]), HALE and ADAMS conclude from their wavelength measurements in the regions λ 4492—λ 4584 and λ 5111—λ 5198 that the chromospheric lines coincide very closely in position with the Fraunhofer lines, as measured by ROWLAND. The average difference — taken without regard to sign — between the wave-lengths of 121 chromospheric lines and those of the corresponding Fraunhofer lines, was found to be only 0.013 Å; and if the sign was retained, the average systematic deviation came out + 0.002 Å, which was much less than the error of measurement.

These provisional results are stated by them to be opposed to my hypothesis, according to which the bright lines of the spectrum of the chromosphere and the flash are mainly due to anomalous refraction of light coming from the photosphere. They say: "If this were the case, however, we should expect their wave-lengths to differ appreciably from their normal values, being somewhat increased on the usual assumption of a density gradient decreasing outward from the surface of the sun."

In order to recall to mind the particulars of that anomalous refraction theory, HALE and ADAMS refer to a paper by HARTMANN (Astron. Nachr. 175, S. 347, 1907). I am under the necessity of also referring, for the same purpose, to the first article in which I started the idea that anomalous dispersion might be an important factor in producing solar phenomena [2]); for HARTMANN, in his criticism, does not do full justice to the argument there developed. Indeed, HARTMANN puts it [3]) as if I had overlooked a simple consequence of my own assumptions, viz. that, if the chromospheric light were due to anomalous refraction in a mass of gas, the density of which gradually decreases outward, only R-light [4]) could appear in the spectrum of the chromosphere. Evidently, however, I was fully aware of that consequence, as this very consideration induced me to think, that to the appearance of the chromosphere and its spectrum the *irregular* anomalous refraction

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[1]) GEORGE E. HALE and WALTER S. ADAMS, Contrib. from the MOUNT WILSON Sol. Obs. No 41; Astroph. Journ. **30**, Oct. 1909.

[2]) JULIUS, Proc. Roy. Acad. Amst. II, p. 575 (1900); Astron. Nachr. **153**, S. 433 (1900).

[3]) HARTMANN, Astron. Nachr. **175**, S. 349 (1907).

[4]) By R-light and V-light we indicate waves, lying very near an absorption line, on its red and its violet side respectively.

must be material in a much higher degree, than the regular ray-curving round SCHMIDT's critical spheres.

So I imagined the solar atmosphere to be honeycombed with irregular density gradients, which may be steeper than the underlying general radial gradient. The views I then held appear from the following qnotation [1]):

"The light of the chromosphere and of the flash lines may be symmetrically distributed on either side of the corresponding Fraunhofer lines; if so, they seem to coincide with the latter; but in certain places of the limb the case must arise that the bright lines would appear to have shifted their position with regard to the absorption lines. For according to the distribution of the density of the vapours, it will be, in turns, especially the rays with very great refractive index (on the red side of the absorption lines), and those with very small refractive index (on the violet side of them) that are curved towards us.

As, upon the whole, the density of the gases of the solar atmosphere will decrease rather than otherwise in proportion as they are farther from the centre, it may be expected that the bright lines will oftener shift their position with respect to the Fraunhofer lines in the direction of greater wave-lengths than in that of smaller.

These details will probably become clearly visible in the eclipse photograms obtained with slit-spectrographs of great dispersion. It is not impossible that in many of the chromosphere-lines a dark core may be seen."

HARTMANN, on the other hand, says: by anomalous dispersion V-light cannot possibly appear with any appreciable intensity in the chromospheric spectrum. From his argumentation [2]) we must conclude that the scheme of *radial* density variation was present to his mind, but that he did not realize the very probable existence of considerable gradients in other directions.

In this matter HALE and ADAMS seem to share HARTMANN's opinion.

We, therefore, will once more draw attention to the importance of *irregular refraction* at the sun's limb, and inquire whether the provisional results obtained by HALE and ADAMS from their photographs of the flash spectrum are really incompatible with the hypothesis, that the chromospheric light is mainly a refraction phenomenon.

Let our starting-point be the same supposition, which also served as a base in a recently published paper on "regular consequences of

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[1]) Proc. Roy. Acad. Amst II, p. 584–585; Astron Nachr. **153**, S. 442 (1900).

[2]) HARTMANN, l.c.

irregular refraction in the sun" [1]), viz.: that at a certain level of the solar atmosphere (e.g. in the so-called reversing layer, or perhaps lower down) the irregular density gradients are comparable in magnitude with the vertical gradient of our terrestrial atmosphere. By a simple calculation we showed, that under such circumstances even normally refracted waves may be bent into curves whose radii of curvature are many times smaller than the radius of the photosphere. Light, suffering anomalous dispersion, is curved more strongly again. And because the solar atmosphere has a particularly great refracting power for R-light and V-light appertaining to lines of its *principal* constituents, rays of that sort may be quite appreciably curved even in much higher levels, though the density gradients are smaller there.

Now, it is evident that not only R-light but also V-light certainly can appear outside the limb of the solar disk by anomalous refraction.

In the subjoined figures the arcs represent the limit of the photosphere. Let regions where a density gradient is supposed to

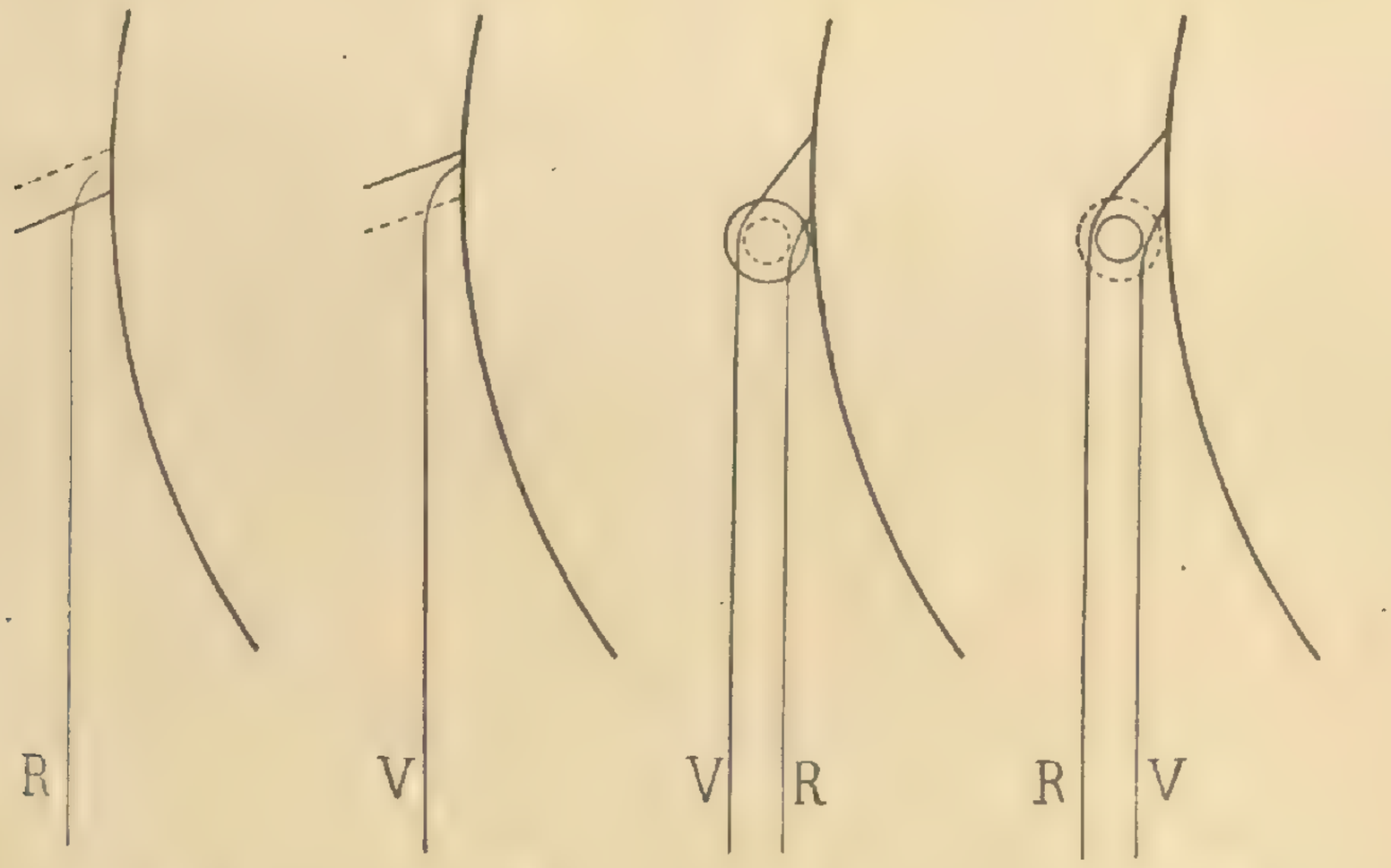

Fig. 1.

exist, be included between a dotted line (where the density be small) and a full line (where the density be great). Figures *a* and *b* thus refer to nearly plane layers with varying density, fig. *c* to a region including a minimum, fig. *d* to a region including a maximum of density. The lines R and V then show us the paths of R-light and V-light.

R-light surely has a better chance to become visible beyond the sun's edge, than V-light, because the general radial gradient, which

[1]) Proc. Roy. Acad. Amsterdam, XII p. 266, 1909.

in our schemes has to be added to the irregular ones, contributes but R-light to the spectrum of the chromosphere. However, we are still perfectly ignorant, as to the proportion existing between the value of the radial gradient and the average value of the irregular gradients at a certain level. Considering the sharp contrasts in the complicated, well-defined structure revealed by the spectroheliograph, and leaving its interpretation aside, we cannot find it improbable that the local density gradients — whether in the mixture or in separate vapours constituting the solar atmosphere — should, as a rule, be steeper than the radial gradient. And if such is the case, the chances of V-light will not be much inferior to those of R-light.

The wave-lengths of the *most* deflectable sorts of light on either side of a narrow absorption line differ only so little from the wave-length of the line itself, that practically a finite limit has not yet been assigned to that difference. In the spectrum those waves must therefore almost coincide with the absorption line. Displacements of the essential part, the "centre of gravity", of a chromospheric line may of course occur, in consequence of the casual distribution of matter favouring at times R-light, then again V-light with the proper incurvation towards the earth; but with the weaker lines such displacements are only slight, because they are restrained to the narrow region of great dispersion anomaly.

The width of the wave-length region, in which a certain line of the solar spectrum produces appreciable anomalous dispersion, depends on the concentration with which the corresponding vibrating system is represented in the parts of the solar atmosphere traversed by the beam under consideration. With most lines that region is narrow. We obtain some idea of the width of it by noticing the widths of the Fraunhofer lines (wings included) of the average solar spectrum. This follows from our thesis that Fraunhofer lines are absorption lines enveloped in dispersion bands [1]). According to FABRY and BUISSON [2]) Fraunhofer lines of intensities 1 to 8 (on ROWLAND's scale) have average widths ranging from 0,07 to 0,16 Å, ; our theory requires the "centres of gravity" of chromospheric lines to be included within these regions (apart from exceptional cases relating to extraordinary local density gradients); so we cannot be surprised at the result, that the average difference between the wave-lengths of chromospheric and corresponding Fraunhofer lines is only $\pm$ 0.013 Å.

I see no reason, therefore, to conclude with HALE and ADAMS, that

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[1]) Proc. Roy. Acad. Amsterdam, XII, p. 280.

[2]) FABRY et BUISSON, Comptes rendus, 28 juin 1909.

the results of their provisional wave-length determinations of chromospheric lines are opposed to a theory, which ascribes the main part of the chromospheric light to anomalous refraction.

The current theory of light foresees anomalous dispersion near every absorption line. Experiment has not, indeed, brought out the phenomenon in its full generality as yet; but HERMANN GEISLER [1]), in a dissertation prepared in Bonn, could already indicate more than 260 lines and bands in the arc spectrum, showing anomalous dispersion.

The great extension of the solar atmosphere certainly is a favourable circumstance for dispersion phenomena to become apparent; in order to produce such phenomena in the laboratory, other favourable circumstances have to be substituted; and if, with certain lines or bands, one does not very quickly succeed in realizing the necessary conditions, it would be rash to conclude that those lines or bands do not give anomalous dispersion.

A limit is set to the possible influence of anomalous refraction on the distribution of the light in the sun's image, by the *scattering*, which a beam of light suffers in any space containing matter, even if the particles are not greater than molecules. In consequence of that scattering the intensity of the direct beam will be reduced to a certain fraction $\left(\text{e.g. } \frac{1}{e}\right)$ of its original value, as soon as the beam has accomplished a certain distance, determined by the average density of the matter in the space traversed, and by the magnitude of the particles. Some obvious effect of ray-curving will result, if the following condition be fulfilled: the radius of curvature of the rays must not be very great as compared with the length of the path on which, by scattering, their intensity is reduced from 1 to $\frac{1}{e}$.

As a matter of course, this limitation of the possible influence of ray-curving on solar phenomena affects in the first place those inferences, which are based on the assumption of *extremely* long distances which the light would have to accomplish nearly parallel to the surface of the photosphere. In view of this fact, SCHMIDT's optical interpretation of the sun's limb will probably have to be modified. On the decidedly shorter paths through the solar atmosphere, however, which, in the case of irregular refraction, have to be consi-

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[1]) HERMANN GEISLER, Zur anomalen Dispersion des Lichtes in Metalldämpfen. Leipzig. Barth, 1909.

dered, the process of scattering by the molecules can only interfere to a much smaller extent with the consequences of anomalous refraction.

Several of the stronger lines of the chromospheric spectrum appear double.[1]) This may be partly due to common reversal by absorption; but ray-curving no doubt contributes appreciably to the effect. Indeed, if we remember that, in the spectrum of the solar disk, the light of the nearest vicinity of the absorption lines appears weakened because a part of it is curved back towards the photosphere, and therefore fails to leave the sun[2]), we easily see that the same thing must hold good for the spectrum of the lower chromosphere. The spectrum of the limb passes quickly but gradually into that of the chromosphere. Those sorts of light, for which the refracting power $(R_m \Delta_m)_R$ or $(R_m \Delta_m)_V$ of the mixture of gases is uncommonly great, have a chance to return from the chromosphere to lower levels; so they are present in the chromospheric light with less intensity than kinds of light for which the refracting power is

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[1]) On the plates, obtained with the prismatic camera by the Dutch Eclipse-expedition of 1901, all chromospheric lines were double. After carefully discussing and examining the circumstances with the other members of the expedition, we could not find an explanation of that phenomenon on the basis of errors in the instruments or in the way of using them; and because in the preceding year I had concluded from my theory that many chromospheric lines must show a dark core (cf. the quotation on p. 447), I unfortunately gave way to the temptation of considering the photographed duplication to represent a real phenomenon, and, therefore, a general property of chromospheric lines (Proc. Roy. Acad. Amst, Vol. IV, p. 195, 1901). On further developing the refraction theory of solar phenomena, however, I found it impossible to explain by it the enigmatical observation, that the distances between the components of those various lines should differ so little in magnitude, and should on an average be as great, as they appeared on the photographs. So I could no longer, in the above result of the eclipse observations, see any special proof of the validity of that refraction theory.

This I wrote to Prof. KAYSER (cf. Handbuch der Spectroscopie IV, S. 595); and it was also communicated in March 1907 by Prof. NIJLAND, in my name, to Prof. J HARTMANN. Evidently, however, the latter did not receive that communication, for in April 1907 (Astron. Nachr. **174**, S. 353) and again in July of the same year (Astron Nachr. **175**. S. 352—353) he criticized the above paper on chromospheric double lines just as if I continued to consider that result of the Dutch expedition as a confirmation of my theory. This, however, not being the case, HARTMANN's criticism has in no way rendered our hypothesis less probable, according to which the chromospheric light is mainly refracted light, coming from the photosphere. HARTMANN's explanation of the double lines appearing on the plates of the Dutch expedition — viz. that they might be due to a slight astigmatism of the prisms — seems to me very probable.

[2]) Proc. Roy. Acad. Amst. Vol. XII, p. 280, 1909.

somewhat smaller, and thus give rise to the dark core in some chromospheric lines.

In explaining further particulars regarding the appearance of chromospheric lines one will have to reckon with the fact, that the refracting power of the solar atmosphere is not symmetrical on both sides of an absorption line (cf. fig. 8, plate I, Proc. Roy. Acad. XII, p. 283), but on an average greater with R-light than with V-light. That asymmetry is in general greater with weak lines than with stronger ones, and must increase when we pass from the red end of the spectrum to the violet end, if the presumption proves correct, that the refracting power $R_0\Delta_0$ of the solar atmosphere for waves not belonging to a region of the spectrum where anomalous dispersion prevails, gradually increases from the red to the violet.

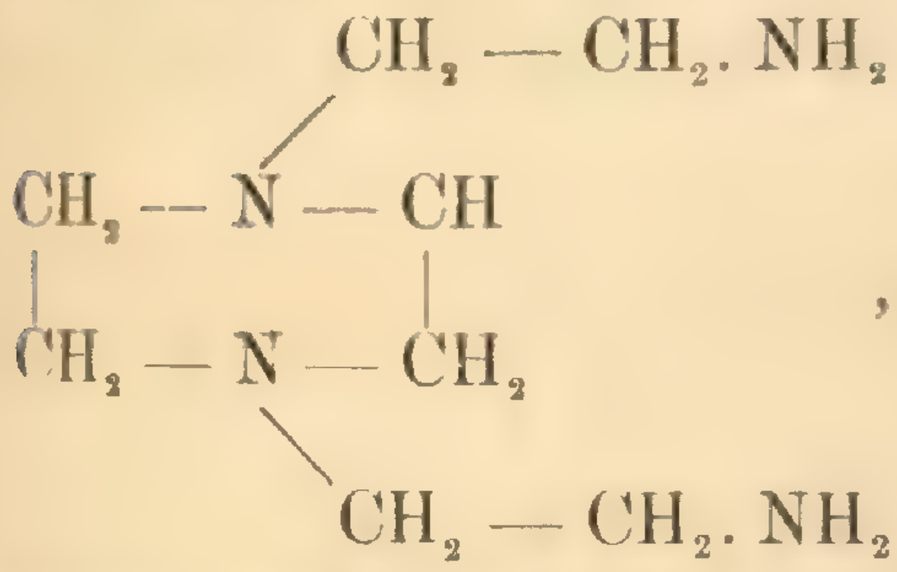
Chemistry. — "*On derivatives of piperazine.*" By Prof. FRANCHIMONT and Dr. E. KRAMER.

In October 1908 Dr. W. A. VAN DORP JR. made known the results of his research on piperazine derivatives. Among other things he had found that by the action of two mols. of formaldehyde and two mols. of hydrogen cyanide on piperazine, *piperazino N(di) acetonitrile* was obtained. From this nitrile, which in aqueous solutions gives no compounds with acids, he prepared the corresponding amide *piperazino N(di) acetamide*, which did combine with acids such as hydrogen chloride (2 mols.), and *piperazino N(di) acetic acid*, which also yielded compounds with acids. He did not succeed in preparing esters of this acid owing to the insolubility of the hydrochloride compound of the acid in alcohol.

We have now succeeded in obtaining the esters of this acid by starting from the compound of the acid with sulphuric acid; this compound contains one mol. of sulphuric acid and two mols. of water to each mol. of piperazino *N(di) acetic acid*. It was heated with an excess of sulphuric acid and alcohol and thus yielded the sulphuric acid compound of the ester, which is crystalline and soluble in water; on treatment with bases, or carbonates under a layer of ether or benzene, the sulphuric acid is abstracted. The *methyl ester* of *piperazino N(di) acetic acid* then consists of a colourless, crystallised substance m.p. 63°, soluble in water, also in ether and benzene. The *ethyl ester* m.p. 47°.5, is also crystalline and readily soluble in water, alcohol, ether, and ligroin and very readily so in benzene and chloroform. Being tertiary amines, both

not only combine with acids but also with methyl iodide but of this they only absorb one molecule; these compounds yield beautiful crystals, that of the methyl ester melts at 144°—145°, that of the ethyl ester at 143°.

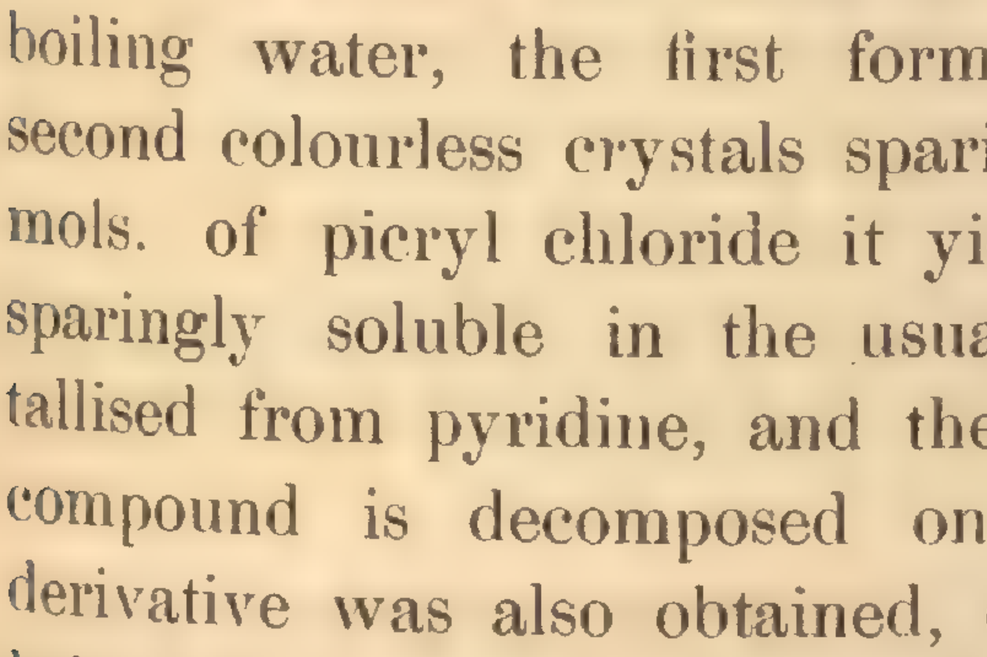
About a year ago I obtained, by the reduction of *piperazino N(di) acetonitrile* with sodium and alcohol a beautifully crystallised amine, which gave the carbylamine reaction and of which the hydrochloride and the hydroplatinichloride compounds pointed, on analysis, to the compound expected, namely *piperazino N(di) ethyleneamine*



, which is, therefore, a cyclic bitertiary amine, and also twice a primary amine, a combination which, as far as I am aware, is not as yet known and is being further investigated by us.

It yields with water a crystallised compound m. p. 63° (the anhydrous compound melts at 40°—41°), which is hygroscopic and gives a white precipitate with NESSLER's reagent. With four mols. of hydrogen chloride a compound is formed which is very soluble in water and yields beautiful anhydrous crystals, which are non-hygroscopic.

Beautiful compounds are also formed with four mols. of picric acid and with four mols. of oxalic acid. When recrystallised from boiling water, the first forms golden yellow lustrous scales, the second colourless crystals sparingly soluble in cold water. With two mols. of picryl chloride it yields a picryl derivative which is very sparingly soluble in the usual organic solvents but may be recrystallised from pyridine, and then forms brownish-yellow crystals; the compound is decomposed on heating at about 238°. A benzoyl derivative was also obtained, of which more later on. As this amine behaved strangely in some respects we thought it desirable to study one of its homologues also, by way of comparison, and chose in the first, place the lower compound *piperazino N(di) methyleneamine*



. We did not succeed in preparing it from

piperazino N(di) acetamide by applying HOFMANN's original method or the modified process and therefore chose another way, namely, the reduction of the corresponding nitrile with sodium and alcohol. This nitrile was still unknown, although we were acquainted with the co-related amide the dicarbamino-piperazine or piperazino N(di) carbonylamide (piperazyldi-urea) and the esters of the correlated acid dicarboxyalkyl-piperazine (piperazyldiurethane). The sought nitrile is N(di) cyanopiperagine or *piperazino N(di) formonitrile*. It was prepared from bromocyanogen and piperazine in aqueous solution with addition of alkali. It is sparingly soluble in cold water, but more readily so than its homologue, and insoluble in ether. It crystallises from alcohol in leaflets m.p. 168°, which shine like mother of pearl; from water it is deposited in long flat crystals with a strong lustre resembling in form the well known Gypsum troins. This nitrile, like its homologue gives, in benzene solution, a white hygroscopic precipitate with dry hydrogen chloride, which is no doubt a combination with HCl.

It does not combine with methyl iodide and (unlike its homologue) not with benzene either. In water, no compound is formed with oxalic acid.

By reduction of this nitrile with sodium and alcohol we have obtained the desired amine, which crystallises beautifully with water. The compounds with hydrogen chloride, picric acid and oxalic acid have been prepared, and also the picryl and benzoyl derivative and will be described later on in the "Recueil des Traveaux chimiques."

We may state here, however, that the amine is not decomposed on boiling with dilute sulphuric acid; in any case it does not yield ammonia and formaldehyde as might have been expected from a derivative of methylenediamine (which it certainly is).

Physics. — "*Some remarks suggested by a paper by* Messrs. TIMMERMANS and KOHNSTAMM." By Mr. J. J. VAN LAAR. (Communicated by Prof. H. A. LORENTZ.)

In these Proceedings Vol. XII, p. 234 in a paper by Messrs. TIMMERMANS and KOHNSTAMM some remarks occur in reference to my former papers on plaitpoint lines etc., which remarks, in my opinion, rest on a misunderstanding.

I hope later on to collect the papers published by me on the subject mentioned in a book, but I may be allowed already now briefly to refute Messrs. T. and K.'s ideas concerning the results obtained by me theoretically.

a. On p. 235 T. and K. call it a "serious objection" to my investigations "that starting from a special case (the supposition $a_{12} = \sqrt{a_1 a_2}$ is probably referred to) they represent it as the general one and *therefore*[1]) must pronounce as abnormal a type occurring for normal substances."

Here I must remark that I have *never* represented the special case $a_{12} = a_1 a_2$ — from which I only started to make the *calculations* feasible — as the general case. Cf. my paper of March 1905 p. 650, where it distinctly says: "The *suppositions*[1]), on which the following calculations are based, are consequently the following." Etc. (Cf. there 3).

Nor have I ever *on account of this supposition* pronounced type II occurring for *normal* substances (fig. 3*a* of the paper of June 1905; afterwards I called this type III) with two cusps R_1 and R'_2 in the plaitpointline (which *also* occurs for $C_2H_6 + C_2H_5OH$, etc.) to be abnormal. I have simply kept the existing nomenclature and spoken of an abnormal *type*, "abnormal", because the plaitpoint line does not run directly from C_1 to C_2 as for type III (fig. 2*a* loc. cit.; later type II), and because it had first been supposed that this so-called "abnormal" type could only occur for abnormal substances.

The result of my observations was exactly that this "abnormal" type might very well occur for *normal* substances.

So far I had not yet got, however, in my paper of May 1905. The investigation begun there was only completed in a subsequent paper. Therefore I only wrote on p. 29 loc. cit. (line 9 from the top): "At all events the anomaly of one of the components *can*[1]) give rise to the occurrence of this second principal type." This refers, of course, to KUENEN's well-known experimental investigation concerning $C_2H_6 + C_2H_5OH$, etc.

But in a later paper (evidently overlooked by T. and K.) viz. in These Proc. Vol. IX Sept. 1906 (The longitudinal plait) I stated on p. 227 as a *result* of my investigations (four papers in these Proc. and two in the Arch. Teyler — see p. 227, line 2—3 from top): "So it appeared that *all* the abnormal cases found by KUENEN may already appear for mixtures of *perfectly normal* substances". (The italics are already found in the quoted place).

This was founded on the investigation, published by me in the Arch. Teyler (2) T. XI première partie, 1902: Les courbes de plissement etc. et sur le pli longitudinal. This paper begins with § 1.

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[1]) The italics are ours.

La possibilité du type III (before called type II by me). On p. 4 of this paper in Teyler in fig. 2 the *coëxistence region* of this type is graphically represented, viz. the region *PBQB*. This figure has been reproduced in the paper in These Proc. of Sept. 1906 (see the Plate, fig. 1). In this paper on p. 225—231 I gave a *recapitulation* of *all* my previous papers. Compare particularly p. 230: "It is now of the greatest importance," etc. "This investigation forms the conclusion of the last paper in the Arch. Teyler." And on p. 231: "In any case the investigations described in the Arch. Teyler have *proved* that this very abnormal type III is *possible for mixtures of normal substances*" etc. (The italics are already found in the original paper).

b. Further T. and K. object to my investigations (p. 235 of their paper) that they, "very onesidedly, lay the stress on the existence of open plaits, a circumstance, which by no means can be considered as a *result*, as it immediately follows from the arbitrary, if not erroneous *supposition* of the linear dependence of b and x."

Now I have never asserted that $\frac{d^2b}{dv^2} = 0$ would always agree with what actually happens; again I have simply *assumed* this, in order to make the *calculations* possible. (See p. 649 of the paper of March 1905 under 2).

c. The remark on p. 235: "For the case at last that one of the components is *abnormal*, VAN LAAR arrives at saturation lines of a very complicated form", etc. — this remarks seems very strange to me. I do not remember having ever theoretically treated the case that one of the components is abnormal. The papers, viz. published by me before the paper of March 1905 cannot be considered as belonging to the series which begins with this paper, in which for the first time the problem of the *plaitpoint lines*, and everything in connection with it, was strictly treated. And I never published anything concerning anomalous substances in this series of papers either. I hope to do this perhaps on a future occasion.

d. In connection with p. 236 of T. and K's paper I only mention that it was by no means generally doubted up to now that for perfectly normal substances *non-miscibility* can occur. VAN DER WAALS himself already stated the conditions of non-miscibility for normal substances in his Continuität II, p. 43. But it was only doubted whether some "abnormal" *forms* of non-miscibility (Type I and III,

occurring among others for $C_2H_6 + CH_3OH, C_2H_5OH$, etc.) could occur for perfectly normal substances.

e. In T. and K's paper it says in a footnote on p. 242 that in later papers I should have left out a dotted line, occurring on the plate of an earlier paper, "*probably* because the special suppositions of VAN LAAR render the occurrence of the required homogeneous double plaitpoints impossible for normal substances."

I have only omitted the case represented by this line because the different course (either to the left or to the right) of the p-T-line was totally unessential for my investigations. But by no means because my special suppositions would render the occurrence of these double plaitpoints impossible for *normal* substances. On the contrary!

f. The inaccurate remark on p. 242 at the bottom of T. and K's paper beginning: "In VAN LAAR's later paper" will not call for any further refutation after the above remarks. Of the *seven* papers published by me on this subject, Mr. T. and Mr. K. seem to have read only two, those of March and May 1905. All the following papers, in which the investigation of the first two was continued, have apparently escaped their notice, particularly the papers mentioned above under *a*.

In conclusion I will only remark that type II has by no means been modified by me, and that I have *proved* (TEYLER loc. cit.) that such an intricate system of plaits is certainly possible on my "*simple*" suppositions, and that I have nowhere thought I had to ascribe the occurrence of this type to *abnormality* (See above sub *a*). On the contrary I asserted the very *opposite*, as clearly appears from the above quotation from my paper of Sept. 1906 These Proc.: "So it appeared" etc. p. 227.

And this, I think, refutes sufficiently all Messrs. TIMMERMANS and KOHNSTAMM's remarks in reference to me.

Baarn, Nov. 15, 1909.

(December 23, 1909).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

APPENDIX

TO THE

PROCEEDINGS OF THE MEETING

of Saturday October 30, 1909.

(Translated from: Bijvoegsel aan het Verslag van de gewone vergadering der Wis- en Natuurkundige afdeeling van Zaterdag 30 October 1909. Dl XVIII).

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Note on some determinations of the lowering of the level of the ground of the Netherlands along the sea coast, deduced from records of water levels and levellings. By Prof. H. G. VAN DE SANDE BAKHUYZEN.

It will only be examined in this note in how far we are justified in concluding to changes of the level of the ground compared with the surface of the sea from old records of water-levels in Amsterdam, and from readings of water-gauges published by Mr. RAMAER, Head-engineer, Director of the hydrographic survey.

1. *Water level of the Y at Amsterdam.*

In a communication in the meeting of the Roy. Acad. of Sciences at Amsterdam of March 1908 (Proceedings Vol. X p. 703), of which a copy is annexed, I concluded that the level of the ground of Amsterdam did not perceptibly sink or rise in relation to the mean

level of the Northsea during the period 1700—1860. A short extract from this communication may follow.

From 1700—1860 the level of the Y at Amsterdam was daily and hourly noted down at the town's tidal station. As appears from these records the difference in height of high water and low water remained unchanged all through this period, from which follows that the communication between the Y and the Northsea has not sensibly changed, and so the results of the changes in the mean level of the Y apply also to the mean level of the Northsea.

From this record I have further derived the most probable level of the water of the Y in the first and the last year of observation, 1700 and 1860, with reference to the zero of Amsterdam (A.P.) at the tidal station, and I have found for it — 165 mm. in 1700 and — 80 mm. in 1860, both with mean errors of $\pm$ 12 mm.

In the years 1860 and 1875 the difference between the height of the zero of Amsterdam at the tidal station and that of the A.P. derived from the grooves in 5 stones, which were placed in 5 sluices in 1682, was determined by accurate levellings. This difference amounted to 82 mm., so that in 1860 the mean level of the Y with relation to the A.P. of the 5 stones was — 80 — 82 or — 162 mm.

Now what was in 1700 the position of the A.P. at the tidal station with respect to the A.P. of the 5 stones?

As far as is known the grooves in the stones placed a few years before 1700 were the only marks by means of which the A.P. was accurately determined, and so it is almost certain that the A.P. at the tidal station has been derived from the height of these grooves by means of simultaneous observations of the height of the water at stagnant water. We may further assume that in that determination a not much greater error has been made than in the determination of the relative heights of the grooves in relation to each other, which probably has taken place in the same way. How accurate those relative heights were then determined, can still be ascertained. In 1875 it appeared namely from an accurate levelling, that the heights of the grooves differed only some mm., at most 8 mm. from each other. It is therefore probable that in 1700 the difference of the A.P. of the sluice stones and the A.P. at the tidal station will have amounted to no more than a few millimeters. The mean height, — 165 mm. of the level of the Y in 1700 with regard to the A.P. at the tidal station may, therefore, also be considered as representing about the mean height with respect to the A.P. of the sluice stones, and this height being — 162 mm. in 1860, we come to the conclusion that from 1700 to 1860 no perceptible change came in the height of the

ground on which the five sluices were built with regard to the mean level of the Northsea. The uncertainty of this result may be expressed by a mean error of ± 2 cm.

If we accept this result, we must also assume that the A.P. at the tidal station has fallen about 80 mm. from 1700 to 1860, but the cause of such a fall is obvious.

The mark had namely been made on a gauging rod, which was thrust into the water through a hole in the floor of the tidal station built over the Y, so far till the cleat at the side of the rod caught on the upper side of this floor. In this position the mark on the rod indicated the A.P., and the level of the water was determined by reading how far above or below the mark the rod was wetted.

It is clear that as well sinking of the floor of the wooden building which rests on poles, as changes of the gauging rod, of the cleat or the mark, which are sure to have occurred in these 160 years, must change the position of the A.P. at the tidal station; and it is certainly less probable that all through this period the assumed A.P. remained unchanged, than that it changed about 8 cm. The improbability that the A.P. at the tidal station should have fallen can, therefore, hardly be adduced as refutation of the above result.

2. *Observations with the water gauges along the seashore.*

Mr. RAMAER, Head-engineer, Director of the hydrographic survey published an important paper in the transactions of the Royal Institute of Engineers in 1908, in which he states the results of an examination of the readings of the water gauges for the determination of changes in the relative level of the coast and the Northsea.

His paper owes its importance among others to the circumstance that these gauges are under the supervision of Mr. RAMAER, who is, therefore, the most competent judge of their reliability. With regard to the reliability I shall, therefore, be entirely guided in my considerations by Mr. RAMAER.

Mr. RAMAER's first calculations are based on readings of 8 gauges placed along the coast, which are evidently considered by him as more accurate than the readings of the other gauges. At least he writes: "We may accept the results found with confidence. This is not the case with the other gauges of table IV (probably table V)." The series of observations used by Mr. RAMAER begins with 1872, because from this year most of these gauges have been regularly observed; it ends in 1906.

Mr. RAMAER supposes the 35 mean yearly sea-levels, which I have represented in their true mutual heights in the adjoined figure, to

be represented by the formula:

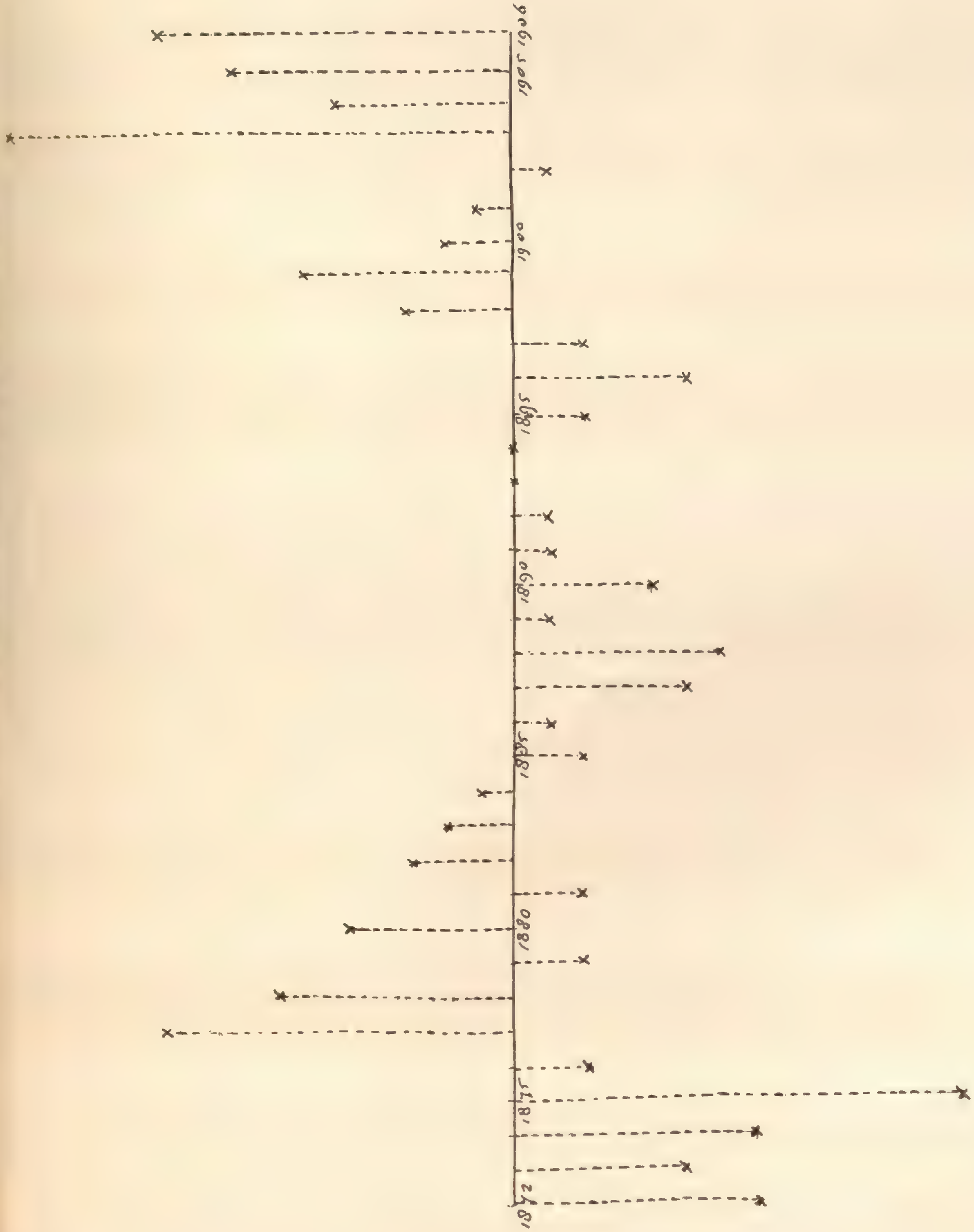
$$y = a + bx,$$

in which x represents the number of years elapsed since an assumed initial date, and b the yearly change. Making use of all these sea-levels, he calculates the value of b by the method of least squares, and finds for it $+ 0{,}13$ cm.; for the mean error of this result I find $\pm 0{,}042$ cm., whereas, if we assume the formula $y = a + bx$ the mean error of a yearly mean is $\pm 2{,}52$ cm.

From the value of the mean error we should conclude that the calculated yearly change, a relative rise of the mean level of the sea of 0,13 cm., is real; from the graphical representation, however this does not appear so clearly, and in order to examine this matter further, I have repeated the calculations of b from part of the data. I have namely excluded the results of the four first and of the four last years, which greatly deviate from the others, as appears from the figure, and determined b from the series of the 27 other years: I find then $b = -0{,}05$ cm., so a relative fall of the sea-level with a mean error of $\pm 0{,}046$ cm. The great difference of the two results, $-0{,}13$ and $+0{,}05$, derived from series of 35 and 27 years, the latter of which is quite contained in the former, shows that only little value is to be attached to those results, and that the sinking of the ground, which is derived from the complete series of observations, is practically only based on the results of 8 years, the first 4 and the last 4, whereas the other 27 years would rather point to a rise.

Beside this first series of readings of the water gauges Mr. RAMAER has dealt with another, which readings were made during the years 1862—1877 on 18 water gauges. (Table XXI of the paper). The 16 mean yearly sea-levels derived from this series he has combined with those derived from the first series of 35 years and he deduced a new value of b from the whole number. It is clear that as in this combination the first series is almost twice as long as the second, the result for b will depend in a great measure on the first found value and cannot be considered as a quite new result. To avoid this difficulty I have calculated the 16 yearly means of the second series separately, and I obtain then for b $+ 0{,}30$ cm., with a mean error of $\pm 0{,}213$ cm. For the mean error of a yearly mean I find $\pm 3{,}93$ cm., from which it appears that Mr. RAMAER justly attaches more importance to the values from the first series which are each based on the mean of 8 water gauges only, than to the values from the second series, in spite of the latter being derived from the mean of 18 water gauges.

H. G. VAN DE SANDE BAKHUYZEN. Note on some determinations of the lowering of the level of the ground of the Netherlands along the seacoast, deduced from records of water levels and levellings.



Yearly means of the heights of the sealevel observed at 8 gauges. Real size.

The new value of the yearly sinking of the level of the ground is not very accurate, as appears from the pretty high value of the mean error. To be able to judge about this in another way I have, just as in the first series, excluded some very divergent values, viz. the yearly means of 1864 and 1877; if we calculate the value of b from the 14 remaining yearly means, we find for it 0.00 cm., in other words: while from 16 yearly means a sinking of the ground of 0,3 cm. would follow, it follows from 14 of the same yearly means that the level of the ground has neither fallen nor risen.

I believe that we are justified in deriving from this investigation that neither from the first nor from the second series of the readings of the gauges communicated by Mr. RAMAER any conclusion may be drawn with reference to a rise or a fall of the level of the ground.

H. G. VAN DE SANDE BAKHUYZEN.

Note on the sinking of the level of the ground in recent years. By H. E. DE BRUYN.

To examine whether of late years according to the observations on the gauges the level of the ground has risen, or whether it has sunk compared with the mean sea-level, the following remarks may be given.

Both in the case that the level of the sea rises, which in my opinion is the most probable case, and in the case that the land sinks, this will not take place regularly, i. e. every year to the same amount, but because nothing is known about this, we can only assume the sinking of the land with respect to the sea-level as regular, so represent the mean level of the sea with respect to the zeros of the gauges as a straight, ascending line.

Now the mean yearly sea-level at any point differs a constant quantity with the height of half-tide. So the rise of the mean sea-level can be derived from the height of half-tide.

The greatest difficulty in these considerations is the possible shifting of the zeros on the gauging rods. As these shiftings are much less frequent for ordinary than for registering gauges I shall only consider ordinary gauges.

The yearly sea-level depends in the first place on the wind. The deviation in consequence of the influence of the wind is great compared with the possible differences in the mean sea-level over some years in consequence of the sinking of the ground. Now it appears from the observations that the influence of the wind is generally in the

same direction during a certain number of years; thus four successive years often occur with high or low level; e. g. 1857—60, 1866—69, 1877—80 and 1903—06.

A comparison of some years, e. g. of 5 years with some other 5 years can consequently give rise to great errors; supposing one period of 5 years is on an average 3 cm. too high through the influence of the wind, the other period of 5 years on an average 3 cm. too low through this influence, this gives a difference of 6 cm.; if now the sinking of the ground during that time is 6 cm., we shall find the sinking of the ground to be $= 0$ if the high years are the first, but $= 12$ cm. if the low years are the first. It is therefore desirable to choose the longest periods possible for a comparison, for the periods in which deviations in consequence of the wind take place in positive or negative sense, may be of pretty long duration. A fixed rule for these deviations or fixed periods for them could not be found. Hence the best thing is to compare the mean of a certain number of years with the mean of another number of years.

A proof that the deviations in consequence of the wind can be in the same sense during many years is furnished in the difference of the mean monthly height of the months inter se, in which all the errors by displacement of the zero point are eliminated. Thus I have found for a 54-year period 1843—1896 of the observations on the gauge at Cuxhaven, that in the first 27 years the mean position in February was 154 mm. higher than the mean position in May, in the last 27 years 43 mm. lower, thus presenting a difference of 197 mm.

In order to derive the probable sinking of the ground from the observation on different gauges, we can, in order to eliminate as much as possible the errors caused by the shifting of the zero points, take the mean of the observations of a certain number of gauges, or draw a conclusion from every gauge and take a mean of these conclusions. For a good survey and comparison, as the deviation in consequence of the wind is not everywhere the same on account of the local conditions, and to enable us to use the observations on that gauge as much as possible for which the longest series is available, the last way is preferable. Of course no gauges are to be used at places where the mean level is modified for some reason or other, as e. g. at Zoutkamp.

When we make a graphical representation of the observations on different gauges, it appears that of late years the sea-level has very *decidedly* risen with regard to the land. The extent of this rise cannot

Year	MUIDEN			BRESKENS	
	Height half-tide mm.	Deviation with a rise of 92 mm. p. c.	Deviation with a rise of 133 mm. p. c.	Height half-tide mm.	Deviation with a rise of 129 mm. p. c.
1855	— 150	— 32	— 18		
56	— 110	7	20		
57	— 190	— 73	— 61		
58	— 175	— 59	— 47		
59	— 145	— 30	— 19		
60	— 145	— 31 — 32	— 20 — 21		
61	— 135	— 22	— 11		
62	— 130	— 18	— 8	— 165	— 23
63	— 125	— 14	— 4	— 170	— 29 — 46
64	— 170	— 60	— 50	— 205	— 65
65	— 135	— 26	— 17	— 205	— 67
66	— 95	13	22	— 90	47
67	— 55	52 — 32	61 — 40	— 90	46 — 38
68	— 80	26	34	— 100	35
69	— 70	36	43	— 110	23
70	— 140	— 35	— 28	— 165	— 33
71	— 135	— 31	— 25	— 150	— 19
72	— 145	— 42 — 34	— 36 — 28	— 140	— 11 — 24
73	— 115	— 13	— 7	— 135	— 7
74	— 135	— 34	— 29	— 150	— 23
75	— 150	— 50	— 45	— 175	— 49
76	— 95	4	9	— 120	4
77	— 25	73	77	— 65	58
78	— 45	52	56	— 80	42
79	— 75	21	25	— 120	0
80	— 40	55 — 30	58 — 33	— 95	24 — 20
81	— 80	14	17	— 120	— 2
82	— 80	14	16	— 100	17
83	— 90	3	4	— 95	20
84	— 60	32	33	— 100	14

Year	MUIDEN.			BRESKENS	
	Height half-tide mm.	Deviation with a rise of 92 mm. p. c.	Deviation with a rise of 133 mm. p. c.	Height half-tide mm.	Deviation with a rise of 129 mm. p. c.
1885	— 95	— 4	— 3	— 115	— 2
86	— 110	— 20	— 20	— 110	1
87	— 95	— 6	— 6	— 130	— 20
88	— 100	— 12	— 12	— 140	— 31
89	— 70	17	16	— 120	— 13
90	— 100	— 14	— 15	— 130	— 24
91	— 95	— 10 — 7	— 11 — 9	— 125	— 20 — 12
92	— 80	4	2	— 115	— 11
93	— 75	8	6	— 105	— 3
94	— 85	— 2	— 5	— 105	— 4
95	— 90	— 8	— 12	— 110	— 10
96	— 100	— 19	— 23	— 115	— 17
97	— 110	— 30	— 34	— 105	— 8
98	— 55	24	19	— 95	1
99	— 60	18	13	— 75	20
1900	— 80	— 3	— 8	— 90	3
01	— 55	21	15	— 100	— 8
02	— 85	— 10 20	— 16 14	— 105	— 14 19
03	— 15	59	53	— 15	74
04	— 65	8	1	— 60	28
05	— 40	32	25	— 55	32
06	— 40	31	24	— 55	31
07	— 100	— 29	— 38	— 90	— 6
08	— 90	— 20	— 29	— 80	3

be accurately ascertained, because the time that we have had sufficiently good observations at our disposal, is too short. A calculation follows here of two gauges leading to an estimation of the extent of this rise. They are the gauges at Muiden and at Breskens. (The values have been borrowed from a paper and an account over 1907 and 1908 by Mr. RAMAER).

Muiden; given the years 1855 till 1908 (this year included).

The rise of the sea-level per century has been derived from the years:

a. 1855—1860 and 1903—1908 (12 years) . . . . . 196 mm.
b. 1861—1902 (42 years) . . . . . 92 „
c. 1855—1865 and 1898—1908 (22 years) . . . . . 196 „
d. 1866—1897 (32 years) . . . . . 17 „
e. 1855—1908 (54 years) . . . . . 133 „

Now the values *a* and *c* are certainly too high, because the years 1855—1865 were low years, and the years 1898—1906 high years. The value *d* is certainly too small, because the years 1866—1869 were high years and the years 1885—1897 low years.

For the assumptions that the rise of the sea-level per century is 92 and 133 mm., we adjoin a list containing the deviations on those assumptions. The mean height of half-tide on Januay 1st 1882 (the middle of the period) is in the first case 94 mm. ÷ N. A. P [1]), derived from 42 years; in the second 96,4 mm. ÷ N. A. P., derived from 54 years. (The height of high water and of low water of every year only being given in cm., a difference of 5 mm. for every year is possible).

It appears from this list that the direction of the deviation for every year is the same in the two assumptions. So this cannot decide in favour of one or the other of the assumptions. It further appears that the periods of permanent deviation either in positive or negative sense may be pretty long, and that the greatest deviations are distributed very irregularly. The most probable assumption is that for so far as displacement of the zero of the gauges has not exercised any influence, the rise lies between 92 and 133 mm. per century.

Breskens; given the years 1862—1908.

The rise of the sea-level per century derived from the 47 years is 129 mm.

In the above-mentioned list the deviation on this assumption has been given. The mean height of half-tide on July 1th 1885 (middle of the period) is 112.6 mm ÷ N.A.P. On the whole the deviations agree pretty well with those at Muiden.

In virtue of the above and because the observations at Muiden and at Breskens are in fairly good harmony with those of other tide-gauges, we may assume as probable that the rise of the sea-level in the last 50 years amounts to about 100 mm. per century.

The Hague, October 1909.

H. E. DE BRUIJN.

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[1]) N. A. P. = Amsterdam zero.

The subsidence of the soil of the Netherlands.

By Prof. G. A. F. Molengraaff.

With regard to a sinking of the soil of the Netherlands one may derive from the latest chapters of the history of geology the following data.

In the tertiary era, probably already in the oligocene period, the subsidence commenced, but it did not affect the whole country to the same extent; on the whole it was more pronounced in the west and the north of the country than in the east and the south, in such a way, that the south-eastern and eastern parts of the country were hardly affected by the movement, if at all, whereas the effect gradually increases towards the north-west.

This is proved by the fact that shore- and shallow-water deposits of the same age formed during that period, are now found in very different altitudes, as appears from the subjoined table compiled from the results of several deep borings. [1])

TABLE I.

	Grave	Goes	Gorkum	Utrecht	Amsterdam
Holocene + Pleistocene	above 4	above 29	above 117.5	above 152	above 200
Amstelien		29—34	117.5—178	152—240	200—235
Waltonien = Scaldisien + Poederlien	4—7.5	34—54.5	not reached	240—268	not reached
Diestien = Gedgravien	7.5—20	54.5—93	„	268 – 365	± 450 [2])
Rupelien		93—220			

Depths in meters below A. P. (Amsterdam level) [3]).

The south-eastern and the eastern portion of the Netherlands remained, however, not entirely unaffected. The country there is

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[1]) See H. G. Jonker List of Literature Verh. Kon. Akad. van Wet. XIII Lorié, No. 680, No. 783, and No. 849, Harmer No. 1004, and No. 1191, particularly Lorié No. 1265. and P. Tesch. Der Niederländische Boden und die Ablagerungen des Rheines und der Maas aus der jüngeren Tertiär- und der älteren Diluvialzeit. Meded. Rijksops. v. Delfstoffen (Communications of the government institute for the geological exploration of the Netherlands) N. 1. 1908. p. 12.

[2]) This depth is not reached in the boring, but the figure 450 is found by calculation.

[3]) The Amsterdam level (A. P.) is considered to be the normal average level of the sea.

broken and faulted; elongated tracts of land which took no part in the downward movement, so-called "horsten", (uplifted blocks, "Horste") alternated with tracts which took part in it, so-called "slenken" (sunken blocks, "Gräben").

The trend of these tracts of ground, whose existence has got known of late years particularly by the deep borings made by the goverment Institute for the geological exploration of the Netherlands is in general northwest-southeast in the south-eastern part of the land.

Moreover it is known that for a great part of the Netherlands this subsidence has never given rise to great differences of level on the surface, because the effect of the sinking was continually more or less counterbalanced by the accumulation of deposits of sand and silt on the sinking blocks. This is proved by the fact that the character of the fauna of these tertiary deposits down to the greatest depths reached until now, continues to be that of a shore or shallow-water fauna.

Without any doubt the depth of the sea was never more than 60—70 meters in spite of the great amount of this subsidence.

We must accordingly assume that in tertiary, especially in late-tertiary time the rivers already carried along large quantities of fine sediments (sand and silt), towards the North sea in our country as it was then, which sediments slowly raised the sinking bottom of the sea to such an extent that the accumulation always kept pace with the subsidence. As the amount of this subsidence increased towards the north-west, the quantity of the sediments deposited in an equal space of time had also to increase in that direction, as clearly appears from table I.

The same thing is observed in the east and the southeast of the country in the sunken tracts "slenken". In a "horst" a certain formation deposited in a certain space of time can be very thin, or may fail altogether, whereas in the adjoining "slenk" the deposits formed in the same space of time may have a considerable thickness as appears from table II [1]). (See p. 470).

Towards the end of the tertiary period the transporting power of the rivers gradually increased, and the amount of sediments deposited was consequently augmented. The subsidence of the soil ceased temporarily, or was possibly interrupted by a feeble movement in the opposite direction. A real or apparent upheaval of the land, at all events a negative shifting of the shore line was the result; the sea was forced back, and the Netherlands became for the greater part, or entirely, dry land.

[1]) See P. Tesch loc. cit. and Verslag der Rijksopsporing van Delfstoffen 1906.

TABLE II.

	Boring Vlodrop 1 in "slenk" South-west of Peelhorst.	Boring Vlodrop 3 in Peelhorst.	Boring 7 in "slenk" North-east of Peelhorst.
Pleistocene	28 + A.P. — 161—A.P.	70.9—65.9 + A.P.	31.9—16.9 + A.P.
Pliocene "kieseloolith stage"	161—532 — A.P.		16.9+A.P.—26.1—A.P.
Pliocene Marine	. . . . .	. . . . .	26.1—118.1 —A.P.
Miocene Lignitiferous sand		65.9+A.P.—171—A.P.	
Miocene Marine			below 118.1 —A.P.
Oligocene		below 17.1 —A.P.	
Thickness Pleistocene	189	5	15
Thickness Pliocene	371	fails	135
Thickness Miocene	not reached	83	

Also a great portion of the North sea must have become dry land in the late pliocene times, so that a branch of the Rhine flowed through the eastern provinces of England, as HARMER has shown. The approach of the glacial epoch announced itself by the increasingly marked boreal character of the fauna of the transition strata between Pliocene and Pleistocene, the Scanien and Weybournien.

In the succeeding diluvial or pleistocene period the soil of the Netherlands resumed its descending movement, but the transporting power of the rivers also increased greatly; besides silt and sand, large quantities of gravel were brought to our country, from which material the so-called gravel-diluvium was formed. And not only the rivers conveyed material, but also the Scandinavian land-ice reached our country in the chief glacial epoch (the Riss glaciation), and covered it with the exception of the portion south of the Rhine. The same thing took place on a much smaller scale according to some scientists in the succeeding last or Baltic glacial epoch. By the pressure of the ice the surface of the gigantic fluviatile sand and gravel-delta over which it moved was greatly altered, and the ice itself left much material of Scandinavian and Baltic origin behind in its ground moraine (the "keileem") and its other moraines, while also the glacier streams modified or covered the pre-glacial stream-deposits both by erosion and by depositing. The country however continued to sink, and the enormous quantities of gravel and sand, carried on by

water and ice, could not do much more than neutralize the influence of the subsidence.

In the interglacial periods with their drier climate the influence of the sinking was decidedly very marked again. Thus LORIÉ [1]) succeeded in showing that, in the interglacial period which succeeded the great (Riss) glacial epoch, thus previous to the last great period of extension of the pleistocene glaciers, the effect of the subsidence was greater than the counterbalancing effect caused by the accumulation of sediments. The northwestern and western part of the Netherlands was then partly flooded by the sea, and in a gulf the strata of the so-called Eem-system, a shallow-water and shore formation were deposited. But then the influence of the sedimentation compared with that of the subsidence became stronger again; riversand again covered the sea-clay of the Eem-system, the sea was forced back again, and rows of dunes originated successively further and further to the West and Northwest far out in the sea as compared with the present shore. But the sinking continued; the strip of land lying inside the dunes was flooded by the sea and covered with water. Holland was for a great part converted into a lagoon or "haff". More than once the water in the lagoon, probably during short intervals of contrary movement, became shallow, brackish and finally fresh enough to be filled up with a peat-vegetation. Then the sinking of the ground increased again, and sea-clay covered the peat once more. Almost everywhere in South-Holland and North-Holland these old peat deposits are found under the blue sea-clay, generally at about 12 m. below A.P., sometimes at greater depth, e.g. at 19 M. below A. P. at IJmuiden, and between Hoorn and Zaandam. Probably the origin of the natural gas, which is used for illuminating purposes in many farmhouses in North-Holland and South-Holland, is to be found in these deep peat-deposits under the sea-clay. Our knowledge, however, of these deep peat-deposits and the natural gas is still very imperfect.

Perhaps it is to be recommended to fix the beginning of the holocene or recent period in the latter part of the lagoon stage. In the first, pre-historic part of the holocene the dunes extended a good distance further sea-ward than now, but the considerable destruction of the coast line by the action of the waves (only after the Pas de Calais was formed, probably at the end of the pleistocene, the destructive effect of the sea became very powerful) made them decrease rapidly, just as is still the case at present. The lagoon was filled with sea-water down to a depth of about 6 meters. As LORIÉ has shown, a short period of upheaval amounting to about 6 meters, followed, which

[1]) See JONKER's list Nº. 725.

was again succeeded by an equally great subsidence. In consequence of this rise the lagoon was almost converted into dry land, the water became fresh, and a peat vegetation filled the lagoon. This peat continued to grow during the slow sinking that followed, till it reached the thickness of ± 6 meters, which it now has.

Now the second, historical part of the holocene follows. The transporting power of the rivers is slight, and modified by man in a sense unfavourable for the accumulation of deposits by canalisation and normalisation of the rivers. The sinking continues and the loss of land is accelerated by the rapid wearing away of the dunes on the sea-side, and more inland by floods caused by storms. A subsidence of the soil in historic times is proved by the situation of structures of Roman origin, as the Brittenburg near Katwijk, and the Nehelennia temple near Domburg, which were still inhabited A. D. 300, as appears from the coins found there, and which therefore cannot have been below A.P. then, whereas now they are resp. 1,5 and 4 m. below that level. For a continuous sinking also in later ages, between 1300 and 1750, urgently pleads also the fact that the polders lie the deeper below A.P. as it is longer since they were drained. This is sometimes ascribed to a prolonged shrinking of the ground, but the conditions which give rise to this phenomenon, are only realised in the upper portion of the ground of the polder which lies now above the artificially lowered level of the groundwater, while it is difficult to imagine dessiccation and shrinking in consequence of it in the deeper lying portions of the ground, which are perfectly saturated with water. Thus it seems very unlikely that shrinking can have given rise to any considerable lowering of the ground, and can have continued for several ages.

The exact amount of the sinking in historic times cannot be determined with perfect certainty, but it may be assumed that it was greater in the north of the country than in the south, wheras the estimations vary between 25 and 90 cm. per century [1]).

If we finally proceed to recent history, say of the last two centuries, it strikes us that investigations about the relative position of the land with regard to the sea-level during that time have led to divergent results.

LORIÉ assumes that the subsidence of the soil of the Netherlands came to a (temporary) stop in the middle of the 16th century, which took place somewhat earlier in Belgium according to RUTOT.

RAMAER [2]) has derived from observations on gauges, carried out

[1]) Cf. JONKER's list No. 313, 782, 785, 810, 913.

[2]) J. C. RAMAER. De daling van den bodem van Nederland. Verh. Kon. Inst ituut van Ingenieurs 1907/08, p. 1.

during the years 1862—1906, that the soil of the Netherlands is still sinking, and that this sinking amounts to about 18 cm. a century.

On the other hand VAN DE SANDE BAKHUYZEN [1]) has derived from observations concerning the level of the official A.P. at Amsterdam, that it remained constant from 1700—1860, and that accordingly the ground of Amsterdam neither rose nor sunk during that period.

The results to which RAMAER and VAN DE SANDE BAKHUYZEN have come, are not very well to be brought to accordance, unless we assume that the sinking set in again after the year 1860 after a period of rest, or as Mr. RAMAER is inclined to believe, that at Amsterdam also previous to the year 1860 the same sinking took place, but that it was just counterbalanced by a movement of the ground in consequence of the upward pressure of the underground marsh-gas. Neither of these explanations appears to be exempt from arbitrariness; for the present, acknowledging the question as an unsettled one, I will choose the case which is the most favourable for the Netherlands, viz. that of stability of the ground during the two centuries preceding the year 1860.

Summarizing the geological history teaches, that at least from the late-tertiary time the ground of the Netherlands has sunk, that this sinking, however, has not been continually perfectly gradual, but that it has now and then been interrupted by shorter of longer periods of rest or movement in an opposite direction. LORIÉ has demonstrated one period of opposed movement with certainty in the lagoon stage while we may conclude to others with great probability on similar grounds. Also it has been concluded from the stability of the level of the A.P. at Amsterdam that there has probably been a period of rest during the two centuries preceding the year 1860.

Before discussing the practical signification of this result it is necessary to inquire into the real nature of this subsidence.

The movement of a block of land with regard to the surface of the sea may be caused either by actual movements of the land or by variations of the height of the sea-level. To evade the difficulty of giving a decision in every case, a neutral nomenclature has been adopted, and we speak of a positive and a negative shifting of the coast-line.

In our case, however, the decision is not difficult.

The sinking of the soil of the Netherlands has been an exclusively local phenomenon as appears from the above tables I and II. At short

[1]) H. G. VAN DE SANDE BAKHUYZEN. "The height of the mean sea-level in the Y before Amsterdam from 1700—1860". These Proc. X p. 703.

distances the differences are very great; since the beginning of the Diestien the ground has e.g. sunk 20 meters at Grave, 92 meters at Goes, 450 meters at Amsterdam. The countries adjoining on the south and the west, Belgium and England, have not taken part in this movement.

So we find a real sinking of the land in the Netherlands, which movement has been confined to a comparatively small region.

What then can be the practical significance of the supposed stability during the 200 years preceding the year 1860 in connection with the gradual considerable sinking in the latest geological periods?

Not one single movement of the crust of the earth takes place uninterruptedly in the same direction without intervals of rest or of movement in the opposite direction. Nor does it here. At least since pliocene times the ground has continually sunk, but no doubt this movement has been interrupted by periods of rest or slight opposed movement, which can be really proved, as was stated above.

But these are only slight sinuosities in a continuous line, whose general direction is downward. We cannot assume on geological grounds, that a movement, which has proceeded regularly in the same direction during a long series of centuries, however irregular it may be if only a short portion of time is considered, should have ceased just now, because we have not succeeded in demonstrating it by measurement during the last two centuries. No, we must much sooner assume that the two centuries before 1860 present one of the numerous small inflections in the line of movement, which on the whole runs downward. The rest will certainly be of short duration, probably it has already been replaced by a descending movement, and geologically speaking we may say in spite of the rest during the 200 years preceding the year 1860 observed at Amsterdam: the ground of the Netherlands has constantly sunk at least since pliocene times, and is still in the same long period of subsidence.

The Hague, June 11th 1909. G. A. F. MOLENGRAAFF.

Note on the sinking of the level of the ground of the Netherlands. By J. M. VAN BEMMELEN.

The depths at which the old layers of peat in the quaternary period of the strata of the earth are found, render the assumption of a lowering of our soil necessary, which is not unimportant. Dr. LORIÉ has derived from several borings that the sinking in that time has at least amounted to 18 meters, for at this depth the layer of peat has been found, though originally it has lain at the surface, and the

pools in which the peat has been formed, and to the bottom of which it has sunk, can have been but a few meters deep as a rule. How long it is, however, since such a peat layer was formed in those pools, is not to be estimated. So we cannot calculate how many years this sinking has continued, or to how much it has amounted in a thousand years or in a century, supposing it to have been regular throughout that time.

If we consult the opinions of geologists on this subject, we see how divergent these opinions are.

Mr. STARING considers the hypothesis of a sinking in recent times improbable, and ascribes the sinking for so far as it has been observed here and there, to the pressure of the woods, which have grown on the layers of peat.

SUES arrives at a negative result for our North-sea and the Baltic-sea shores. He is of opinion that visible sinkings were brought about by landslips, or by ruptures in not embanked places, or by storms, or by climatic changes in the Baltic region.

PENCK does not assume a sinking for the latter part of our present geologic period, and does not admit any measurable shifting of the coastline for the last thousand years.

DE GEER, too, assumes that the beach in our days is comparatively in a period of rest.

GEINITZ adduces some new facts, it is true, which point to a sinking, and mentions the formation of the Zuiderzee and the bay of the Dollard in the Netherlands, of the Jahde bay in North-Germany, the tradition of the Cimbric flood, but does not consider them as certain proofs.

SCHUCHT accepts, indeed, a secular lowering at the time of the depositing of the layers of old-alluvial silt and the formation of marshes (e. g. of the Weser), but is of opinion that no lowering can be ascertained at the time in which the Dutch range of dunes was broken and the conditions of ebb and flood were modified on the Northsea-coast.

SCHÜTTE thinks that the fact that the seaclay looses its chalk in course of time and the silt-layers shrink by desiccation and compression in consequence of the weight of the new silt-layers, could bring about at the utmost a time of rest in the lowering. Formerly it is true, he thought he could accept a lowering of 7,5 mm. a year (so 7,5 m. every 1000 years). Afterwards, however, he has revised his opinion, and has rejected the grounds for it.

Dr. REYNOLD estimated the sinking of the ground of the East-Frisian Dollard-polders at 8,8 mm. a year, and VENEMA [1]) has thought

[1]) VENEMA. Over het dalen van de noordkuststreken van ons land.

he could derive from the present height of the Groningen Dollard-polders, a sinking of 8 mm. a year. If it was allowed to apply that cypher to a longer period, the sinking would amount to 2—2,2 m., in the time from 1545 (the year of the first embankment) till 1850 and of 8 –9 meters in 1000 years. This estimation exceeds so much the estimations derived from other data that for this very reason it appears to be unacceptable.

We bear in mind, however, that VENEMA's values also comprise the sinking of the ground of the Dollard-polders in consequence of the shrinking of the clay and peat layers during those 200—300 years. Beneath the clay the rests of the layer of peat remain, of which the ground consisted in earlier times and on which the Dollard-clay has settled. How thick these rests are (probably 1—2 m.) and to how much the shrinking in consequence of the deposited clay has amounted, is unknown, and so no estimation is possible.

In 1872 Dr. G. A. VAN GEYTENBEEK [1]) made a study of the situation of the layer of peat or of "derrie" (clayey peat) under the clay and of the layer of clay in the Zealand polders. He found that the upper surface of the layer of "derrie" lay at about 2 m. below A.P. From the height of the surface of the polders diked in between the 10th and 19th century with regard to the open sea, he derives a sinking of the surface of 2,865 m., so on an average 2,9 mm. a year.

The Insp.-Gen. A. A. BEKAAR has (according to the paper by Mr. J. C. RAMAER, Chief.-Engineer, made this calculation from the data of two hundred polders, and found a sinking of the surface of 1.8 m., so about equally large. This sinking, however, cannot be considered as a sinking of the ground, it is only a sinking of the surface of the polders. Two factors come into play, first the shrinking of the clay by compression, secondly the actual lowering of the ground. From the height of the polders during the successive embankments from the eleventh century up to the present time the amount of this shrinking may be roughly estimated; probably about 1 m. On this supposition the actual lowering of the ground in Zealand would have amounted to only 0,8 m., so not quite one meter.

The old remains on the beach at Domburg (namely the temple of Nehellennia, coffins of a cemetary and many dwellings) lay behind the range of dunes in earlier times. Now they become visible on the beach, and have often been visible since 1680, when the wind continues to blow from the East or Northeast for a considerable time

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[1]) Proeve eener Geologische Verh. over de prov. Zeeland gedurende het hedendaagsche tijdvak.

and the water at ebb has been very low. Thus the self-registering gauges at Westkapelle on the beach indicated the positions:

from 1877—1880	2.68 m.	below A.P.
1881—1890	2.95 „	„ „
1891—1900	2.81 „	„ „

On the beach at Katwijk the low-water-mark is —1.84. There the ruins of the "Brittenburg" became visible in the 18th century.

These remains date from the beginning of our era, and would therefore point to a sinking of from 1,5 to 3 meters, unless the situation should be modified by other causes.

From these observations it might be deduced that the beach at Domburg lies from 2—3 m. below A.P. This result is one of the principal which have led to the opinion that a not inconsiderable lowering of our ground has taken place on the west coast of our country since the beginning of our era.

There are, however, many reasons for attributing this fact to other causes.

Since that time dunes have been blown inland over the beach, so that they lay more westward in earlier times. According to the map published by N. Visscher in 1670 there were still 4 or 5 ranges of dunes on the present beach of Walcheren, and now only one narrow range is left. So the others were washed away or blown inland. There was great pressure of these dunes on the beach at one time, but this has now ceased. The soil, there, consists of clay (which now presents a blue colour). This clay rests on a layer of peat.

Even now detached pieces of peat are found on this beach. This layer of clay is the "kwelder" an alluvial deposit outside the dike on which as on the Zealand islands in general (and also in Friesland and Groningen) the clay sediments were deposited, and on which the Frisian and Groningen Mounds ("Terpen") "terps" and the Zealand Mounds (hillocks "Hills") were erected.

Now there is a picture extant of the beach, made in the year 1860, when it was again quite laid bare by the winds, and the remains of the habitation were clearly visible. This picture shows that the coffins of the old burial ground from the first centuries after Christ lie on the beach; so the sand, in which they have lain, has been washed away. Yet they must have been at some depth *under* the earth of the burial ground, and not uncovered on the bare strand.

As now the dunes have passed over them, they were exposed to a pressure at one time. It is, however, the question if the clay soil was

softer then. It may therefore be questioned if the upper soil was peaty then, and if the oldest inhabitants of this coast-settlement have settled on peat. This, however, is not probable, but the clay layer *rested* on peat at all events, for the fen layer is found in Walcheren at ± 2 m. below A. P.

It is, therefore, of much importance to investigate at what depth this clay and this peat layer of the Domburg beach lie under the dunes, and how thick they are there. This knowledge is still wanting. Till then we shall have to suspend our decision as to the sinking, for not till then can we get a fixed point for the degree of compression of these layers. At present there is still too much discrepancy between the accounts of the situation of the foundation of the finds, the situation of the clay, and the situation of the peat at Domburg.

So we cannot yet decide how much less than 2—3 m. the compression of the ground, and therefore the sinking can have been at Domburg, and if a sinking of any importance remains after that.

It would be of importance to know in what century or centuries the shifting and narrowing of the dunes from the outside inland took place on the western coast of Walcheren to such an extent that some ranges of dunes have disappeared and the place (at earlier times behind the dunes) where the temple of Nehellennia, the burial ground with coffins and the dwelling-places stood, has become a beach covered with sand. But this time is not accurately known. Yet we should have to know it, to be able to conclude when it first became possible that this place was sometimes laid bare with continued east wind, and with very low ebb, and revealed itself with its remains to the people on the beach, as happened for the first time in the beginning of 1647, and has happened later on, even up to the present time [1]).

This cannot have been much before 1646, for on N. Visscher's map of 1670, some ranges of dunes are drawn, which have now disappeared from the map of Walcheren, so that only one narrow line is to be seen. In 1647, however, the displacement and removing of the dunes by the wind must have been in full progress, and even far advanced, else this place could not have become a beach. Before 1647 there can have been hardly any chance of these remains being laid bare, because some dunes still rested on them; else the inhabitants of Dom-

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[1]) Observed in 1687, 1705, 1706, 1707, 1749, 1817, 1832, 1835, 1860, 1850—1866, 1879, according to Dr. Pleyte, and observed 1906 by Miss Marie de Man, who then clearly distinguished the foundation of a house, consisting of square pieces of clay, which lay regularly side by side with a pole in the middle, and with traces of a fence of paling.

burg would certainly have destroyed these remains, as they did afterwards.

So this displacement of the dunes inland must have taken place between 800—1000 before Christ and 1647. During the first centuries after Christ the coast immediately behind the dunes was still inhabited. But because the dunes were blown inland, this coast was deserted for so far as it became beach, and the inhabitants moved to those parts of the island which had been raised and made habitable by deposition of silt. Yet the coast, which had partly become beach, continued to be used, for some years after 1647 the remains of a canal were discovered, which was used by the boats of the herring fishery, and is reproduced by Mr. FREDERIKS on his map of 1866. [1]) It is known that Domburg had still a harbour in 1544, from which the boats sailed out for herring fishing.

So there is no reason to assume that first a sinking should have taken place between the beginning of our era and the tenth century, and still less that a rise should have taken place between the 10th century and the seventeenth century, which caused the remains of the temple, the burial ground, and the dwellings from the Roman time to reappear. A rise has not been necessary to bring this about.

J. C. RAMAER, head-engineer of the hydrographic survey, has derived 1st a sinking of 1,3 mm. a year from the water levels observed on 8 accurate gauges during 35 years, 2nd a sinking of 1.6 mm. from the observations of a series of gauges in our country during 47 years, 3rd a sinking of 1.4 mm. a year from observations during 6 years.

H. G. VAN DE SANDE BAKHUYZEN has subjected these records of RAMAER'S gauges to a strict critical examination, and has arrived at the conclusion, that no reliable results are to be derived from them.

It seems to me that the Mounds in Friesland and Groningen, and also those in East-Friesland, Oldenburg and Holstein might teach us something about the sinking, if we examine the bottom or the floor (of the mound or hillock) (Wierde, Word or Wurth) with respect to the mean high water mark of A. P. For it may be assumed that the erection of the Mound or Wierde has taken place on a "kwelder" [2]) (Schor, Groden) which was dry at low water, and gave protection from the high water by the raised Mounds, as PLINIUS

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[1]) Miss MARIE DE MAN: Que sait-on de la plage de Dombourg? 1899 G. TH. BOM EN ZOON. P. 51.

[2]) A "kwelder" is a new alluvial deposit of clay-silt outside the last dike, or outside the coast.

already described accurately [1]). So the knowledge of the elevation of the floor above A. P. must be of much importance.

For Friesland and Groningen Mr. VAN GIFFEN has collected data about this, and for Oldenburg Prof. J. MARTIN (at Oldenburg). From this appears that for the north of the Netherlands no greater sinking than $^1/_2$ — $\pm$ 1 m. can be derived from these data, nay for many Mounds a sinking which is hardly appreciable, or no sinking at all.

In the first half of the nineteenth century about 1845 Dr. ACKER STRATINGH and Dr. WESTERHOFF stated about the Mounds at Warffum, Uskwerd, and about the Groningen Mounds in general that their bottom lay from two to three meters below the level of the arable ground. The height of the arable ground round the Mound with respect to A. P. was, however, not given, and in no case ascertained by levelling.

Moreover ACKER STRATINGH and WESTERHOFF have very probably set down the foundation (the floor or bottom) of the Mound as too low. This foundation could be most easily ascertained between the layer of manure at the bottom of the Mound and the untouched ground. But it is just at these places that the floor probably lies deeper, for the manure at the bottom of the Mound often lies in dug holes. Such holes have, e.g., been found in the Mounds of Baaijum and of Dongjum. There the remains are found of fresh water basins overgrown with rushes and filled with manure. These basins must have been dug in the earth at the building of the Mounds. When later on the Mound was made higher (which took place with all of them), they were filled up with the manure of the cattle-keepers, as still happens in the Halligen. [2])

We can now by no means derive the depth of the floor of the Mounds, and much less the extent of the lowering, from the accounts of STRATINGH and WESTERHOFF.

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[1]) Cf. "Beschrijving over het tegenwoordig standpunt onzer kennis van de Nederl. Terpen". E. J. BRILL Leiden 1908 p. 144—145.

[2]) The Halligen are oblong, unembanked islands ("kweiders"), now inhabited by cattle-keepers, who do not occupy themselves with agriculture. They were raised high enough by washed ashore silt and mud in former times to be protected from the flood and to be habitable. They lie along the Sleswick coast. On these islands "wurthen" have been raised. If one digs down to the bottom of the Mound one finds the remains of a fresh water basin (Fething) as described above overgrown with rushes and filled with manure. Why and when this happened, is unknown to me. These Wurthen or Mounds date from the preceding century, so they are of recent date.

Some fifteen Mounds in Friesland and Groningen have been examined by Mr. VAN GIFFEN.

Hoogebeintum. On the Mound lies the churchyard, which is very high in itself, and on this the church. The height of the real Mound is 5.80 m. + A.P. north-west of the church. [1]) The Mound must be 1500 years old at least, and the church not less than 700 years, for it was originally built of Tuff-stone, and not before 1200 after Christ bricks came into use as building material, of which the newer parts of the church are composed. The Mound is one of the highest, so we may safely assume that it cannot have been higher after 800. The boundary of the grave-yard (found in the Mound) lies at the edge of the Mound, at $\pm$ $^1/_2$ m. above A.P., and as the relics of this grave-yard date from the Merovingian time of 500—750 after Christ, this grave-yard can be no older than 1500 years. Now the Mound-earth of this Mound does not go any deeper than $\pm$ 0,3 m. above A.P.

The foundation of this Mound consists partly of sandbanks in layers running from the north to the south, and which have been washed ashore from the north-west. The height of these banks (which clearly show "kwelder" vegetation) is from 0,10 to 0,60 m. + A.P.

If the lowering of the ground under the Mound had only amounted to 1 mm. a year, it would have amounted to as much as 1,5 m. after 1500 years, and the graveyard would now lie deeply buried in the ground.

In general the relics from the first centuries after Christ occur in the oldest Mound-layers, especially those from the Roman era, so from the latter half of the first century *before* Christ, e.g, in the Mound at Besseburen near Beetgum, and also the Saxon urns from the Merovingian period. These lowest Mound layers do not lie below A.P. or only a little below it.

The Mound at Baayum. To determine the foundation, the ground close by the side of the Mound was investigated. Under the turf a layer of "knik" [2]) was found, which downward gradually passed into light yellow sandy soil in layers, and under this bluish sand with shells, Scrobicularia, Carduum, Tellina etc.

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[1]) The threshold height of the church is 8.50 m. + A.P.; the highest point of the church-yard lies at 8.95 M. + A.P. This point was formerly erroneously taken for the height of the Mound, which was accordingly given as much too high.

[2]) "Knik" is a clay hardened by mixture with oxyde of iron, and a moist situation during a long time.

Under the Mound the condition was for the greater part the same as in the arable ground. The floor from 0,10 to 0,30 m. + A.P. consists of layers of sandy clay. Some parts of this foundation lay on the North- and on the North-west side. In the basins rush-peat was formed, namely a layer ± 0,85 m. thick. They point to places of excavation for the acquisition of ground for the older Mound. In later times this Mound was made higher, and extended over the original hollows, in which peat had been formed in the meantime. This is the more probable, as no peat is found in the arable ground. The peat mentioned has been greatly compressed by the pressure of the Mound, and yet it lies but little below A.P.

That the foundation of this Mound had not sunk was also derived from this, that under the second surface of habitation, so from the first time of the Mound, shards of *Roman* crockery were found. In the lowest layer of the Mound lay the manure; in the middle of the Mound it passed downward into deep holes, where no manure was found, and the earth had a dark colour, probably in consequence of the oldest habitation. On this different layers, with which the height of the Mound was successively increased, were found, separated by layers of ashes. These layers of ashes mark the different times the Mound was made higher, and each of them we may assume to have been the temporary surface of habitation of the Mound.

The foundations of 7 Groningen and 9 Frisian Mounds were found at a height from just below to just above A. P.

Joeswerd 0,2 above to slightly below A. P.
Dorkwerd at 0.45 m. below A. P.
Oostum at 0.15—0.3 m. + A. P.
Feerwerd at 0.1—05 m. + A. P.
Garnwerd at 0.15—0.5 m. + A. P.
Leermens not considerably below the arable ground.
Westeremden not considerably below the arable ground.
Dongjum at ± A. P. to 0.2 m. + A. P.
Fatum at 0.025—0.26 m. + A. P.
Vlaeren at 0.3 m. + A. P.
Aalsum at the height of the arable ground.
Ferwerd 0.30 — A. P. to 0.05 + A. P.
Pingjum (the village) slightly above A. P.
Terp near Pingjum height of the arable ground.
Kimswerd about the height of the arable ground.
Marsum the same.

Oberahn near Sande in the bay of Jahde lies 1.0 m. above A. P. In the Mound at Farmsum the foundation lay 0.65 m. below A.P., but it rested on 0.4 m. of clay and almost 4 m. of løw peaty ground. So it is clear that the situation of the foundation does not betray a sinking, but a shrinking of the thick peat-layer.

Two Frisian Mounds, which rested on diluvial sand, presented a succession of layers, which pointed to a post-diluvial lowering, so a lowering of earlier times, *before* the erection of the Mound.

On the diluvial sand, (brown on the upper side), followed:

I. A layer of high peaty ground, which contains "rietstokken" (reeds intercrossed with roots) in the lower parts, higher however it contains heather, peat moss, birchwood etc.

2. On this a layer of clay with "kwelder" vegetation. This layer is 1,5—2.0 m. thick under the Mound of Dokkum. In the lowest parts it contains remains of Triglochin maritimum.

So the original surface was diluvial sand.

But since the ground sank and was flooded, so that a vegetation arose in water, which gradually was decomposed and formed a layer of low peaty ground. The sinking of the ground continued, so that clay was washed ashore and deposited by the sea-water. On this sea-clay the Mound was afterwards built. Together the layers are ±3 m. thick near Dokkum, and to this cypher must amount the post-diluvial sinking, which extended over thousands of years, for the above-mentioned Mounds have taught us that no sinking has taken place for the last two thousand years, or only an insignificant one. After the deposition of the sea-elay the post-diluvial sinking must have ceased, or at least have become very slight.

J. Martin furnished some data on the Mounds in Oldenburg.

In the Mound at Bant the floor lies 0,3 m. below the arable land, and cannot have been much higher; for the "kwelder" outside the dike does not lie higher. The ground was rediked in 1529.

The Mound between Ellerdem and Sande, on which a church used to stand in former times, has a recognisable floor, i. e. a sand-layer, which includes a layer of bog, and lies 0.3 m. below the arable ground.

The Mound at Haddien in Jeverland contains a layer of knik-clay which contains many shards down to 1,06 m., under which, sharply defined, a bog layer from 1,1 to 1,5 m. Here the floor lies about at the same level as A.P. The Mound contains many urns with burned remains, hence it dates from the pre-Frankish time.

A Mound at Tettens has a floor which lies 1.2 m. below the level of the "kwelder" outside the dike (= 0,30 above A.P.).

A graveyard of urns, in which many coffins lie, showing the shape of trees, presents an "Oer [1]) soil" of pure gray clay. It is noteworthy that the younger tree-coffins lie deeper. The older urns with burned remains, which were found in it, lie close to the surface and date from the 3rd century.

So if these Mounds in Oldenburg show a sinking of the ground, it is certainly smaller than 1 m.

If we now summarize the results of the investigations, they teach us that:

Some Mounds have shown a post-glacial sinking of $\pm$ 3 m., which took place before the beginning of our era.

The calculations about the lowering of the beach at Domburg are still quite uncertain, because this situation depends on the influence of different other factors. The same thing applies to the beach at Katwijk.

In Zealand the situation of the basis or floor of the Mounds teaches us but little about the lowering. The floors in Walcheren lie from 0.10 m. — A. P. to $\pm$ 0.80 m. + A. P., certainly to 0.50 m. + A. P. They only allow the assumption of a slight lowering. REYNOLD and VENEMA's calculations on the lowering of the Dollard-polders must be rejected.

It follows from the heights of the floors in the Mounds of Friesland, Groningen, and Oldenburg, that the lowering is only a few decimeters, or that there is none at all.

A lowering in the Mound of Farmsum of 0.65 m. may be ascribed to a shrinking of a thick layer of peat under the Mound.

From the data of the gauges at Amsterdam and elsewhere in the Netherlands RAMAER has derived a lowering of $\pm$ 1.5 mm. a year during the preceding century, but according to VAN DE SANDE BAKHUYZEN *no reliable result* can be derived from these data about the lowering of the ground in the last hundred years.

We must further call attention to the uncertainty that prevails as to whether the sinking of the ground in the north of the Netherlands has been the same as in the south, and in the east the same as in the west.

But on this point we have not yet any data laying claim to the least certainty with regard to Zealand, South- and North Holland, Friesland, Groningen, East-Friesland, Oldenburg, Holstein and Sleswick, which would point to this.

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[1]) "Oer" is a layer of sand, containing still more oxyde of iron than the "knik" soil, and hardened by it.

However this may be, this is certain that it has been derived from the geological observations that in the quaternary period there must have been a lowering of the ground of at least 18 m. A determination of the duration of this period is, however, not possible.

In virtue of this, and of a post-glacial lowering before the beginning of our era, which may perhaps be derived from some Mounds, we may state, *that we are still in a period of sinking*, but also that *most probably this sinking has ceased for the last two thousand years*, or at least been very slight during this time.

Zoology. — *"On the Age of Salmon to be Derived from the Structure of the Scales"*. By Dr. P. P. C. Hoek.

(Communicated in the meeting of November 27, 1909.)

Quite independent of human influence, salmon ascend the rivers they think fit in numbers which vary very much periodically. The influence exercised by man — be it men fishing, or men inhabiting and polluting the banks, or men as navigators or river engineers — interferes greatly with the natural production; it makes it difficult, in fact nearly impossible, to obtain an exact notion regarding the extent, the periodicity etc. of the natural fluctuations. Yet the correctness of the ascent varying periodically is not to be concealed by this circumstance: we see that groups of richer years alternate in a very striking way with groups of years of a considerable smaller catch; and they do so at present as well as formerly, judging from statistics at our disposal, say 30 or 50 years ago.

Now seeing how the catches are composed, we soon discover that there is a good reason for this being the case. If we analyse the catch of a certain year we find that considerable differences in size occur between the specimens composing that catch. However, not all possible sizes between the smallest and the largest specimens are represented; on the contrary, according to their size the ascending salmon can be rather easily divided into three categories or size-groups of about the following dimensions:

small	salmon ("grilse" = Jacob-salmon)	of 55—67 cm.
middle-sized	,, (small summer-salmon)	,, 78—90 ,,
and large	,, (large summer-salmon and winter-salmon)	,, 100—110 ,,

Based on a long experience and on a careful study of the sizes of the ascending salmon, which was begun by the Basel Professor Miescher Ruesch (1880) and continued by the present author (1894), the opinion is now well established that the smallest salmon of a certain year and the middle-sized ones of the next year, as well

as the still larger ones of a later year, belong together, which means that they date, if not without exceptions, to a large extent from the same year of birth. Quite in accordance herewith we see fairly regularly, that a year with richer catches of grilse is succeeded by a year with more middle-sized salmon, the following year showing, not quite so regularly but in many instances distinctly enough, that it is still under the same favourable influence. Though exceptions — cases which do not enter so well into the scheme — occur, there is no longer any doubt as to this rule holding good in general. Hence a richer catch of grilse in a certain year has a prophetic significance for following years: it opens a period of years with rising figures of the catches. Rich years, therefore, are not isolated occurrences but form groups alternating with others of poorer years.

So we understand that a good grilse-year is considered by the fishermen also as a good omen for the future. Hence the study of the whereabouts, of the scarceness or abundance in certain years, of these smallest of our salmon has always had a great attraction for me. Questions like the following: why some salmon enter the river as grilse and others not, what the age of a grilse is when leaving the sea and entering one of our estuaries, are to be answered before it will be possible to have an exact notion of the circumstances which are of influence, of the conditions which determine the production of grilse and so of salmon in general. This shows sufficiently, I think, the great importance of determining the age of these salmon; that determination, however, is singularly complicated by the very special way of living of the salmon, its being born in the upper parts of the river, its passing its youth there, its descending to the sea and returning at different ages — a chapter of natural history which I accept to be generally understood by this time, to which, nevertheless, these studies of the age based on the construction of the scale may add a page of some importance.

A few words on the determination of that age in general may precede. To that purpose the length of the fishes is often made use of and this has been done and apparently with some success for the salmon also. However, difficulties arose: a very unequal growth was found in young salmon reared in hatcheries under evidently quite corresponding circumstances: fishes exactly the same number of months old may be 5, 7 or 9 cm. long, and young salmon which have grown up in a free condition may show similar differences in size; of course it is very difficult to reckon with such differences when the age is to be derived from the size. A second difficulty is

of a quite different nature: it is easy enough to catch samples of young salmon so long as they remain in the brooks and smaller affluents of the upper course of the river and so it is possible in the consecutive months to compare these with one another as to their size and to make out their growth; but between the largest that are still observed in the river, and the smallest returning from the sea the difference in size is at once so great, that it would be a hazardous undertaking to derive from it their difference in age. The smallest salmon (smolts) going to the sea and at that time about 14 months old, have a length of 12—16 cm.; along with them descend larger ones of a length of 15—20 cm. and about 26 months old; moreover stragglers are observed in different seasons and at various places in the upper course of the river, which have a length of 18—24 cm. and which most probably are in their third year. The smallest salmon that return from the sea (the so-called grilse) are, however, fishes of a length of 55—67 cm. and salmon between that size and the largest of those descending to the sea in reality have never been observed. Hence any definite rate is wanting when we try to estimate the age of the grilse by comparing their length with that even of the largest salmon descending to the sea.

Following the instructive example given in recent years for other species of fish (especially for the carp [1]) among the freshwater-fishes and for the Gadoids [2]) among the seafishes), of late a beginning has been made to use the structure of the scales for the determination of the age of the salmon [3]). The scales, once formed, as a rule remain with the fish during its whole life; so their number does not increase with the growth of the fish, but, on the other hand, their size does and in a very conspicuous way. The shape of the scales of the salmon is rather regular: in the beginning more oval, later nearly circular. They cover one another like roof-tiles; being placed in oblique rows, only the hindermost part of each scale emerges freely from

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[1]) C. Hoffbauer, Weitere Beiträge zur Alters- und Wachstumsbestimmung der Fische, spez. des Karpfens. Zeitschr. f. Fischerei. XII, 1905. p. 111.

[2]) J. Stuart Thomson, The Periodic Growth of Scales in Gadidae as an Index of Age. Plymouth Journal. VII, 1904. p. 1.

[3]) H. W. Johnston, The Scales of Tay Salmon as indicative of Age, Growth and Spawning Habit. p. 63—79. 1 & 6. Pl. Appendix II to Part II of 23rd Annual Report of the Fishery Board for Scotland (for 1904). 1905.

— The Scales of Salmon. p. 54—66. 5 Pl. Appendix II to Part II of the 25th Annual Report of the Fishery Board for Scotland (for 1906). 1907.

Artur Hutton, Salmon Scales etc. London, 1909; translated in: Allgemeine Fischerei-Zeiting. XXXIV, 1909. p. 459.

the scales placed above and in front of it, the anterior part being totally covered by the adjoining scales [1]). Lines running parallel to its circumference are seen on the exterior surface of the scale; they can easily be made out (even when only a weak magnifying power is used) on the anterior concealed part of the scale and are indistinct or wanting on its posterior area. By developing these lines which are really ridges, the outer surface becomes rough like that of a minute file, and this we can feel when going over it with a needle [2]). These ridges represent so many stages in the growth of the scale. Why it is that the scales do not grow continuously, so to say quite regularly, I do not know; in fact the impression we have of the growth is, that in the course of a year a certain number, 12, 15, 20 new growths, like so many rings, are added to the circumference of the scale and that each growth, in the shape of a delicate ridge, leaves a lasting token on the surface. If the growth takes place more rapidly, then the number of ridges added in the same period is larger; moreover the width of the rings, which is to be judged from the distance of the ridges, increases with the age of the fishes.

If the growth of the scale took place quite 'equally or even with a gradually increasing rate the whole year round, the growth-ridges of the outer surface would not give us better information, with regard to the age of the fishes than the growth of the scale itself would afford. In that case we should only be able to say: larger fishes have larger scales, larger scales have more numerous lines of growth and when approaching the outer circumference these lines are more distant from one another than those of the central part. As, however, the number of rings formed annually is found to be very unstable, it would be impossible in that case to draw conclusions regarding the age of the scale (and the fish itself in general) from the structure of that scale. That this is indeed possible is caused by the following:

1st. because the young salmon, living in fresh water does not take any food during a certain period in wintertime, and does not grow; in that period the scale does not grow either; when afterwards the same fish begins to grow again, this happens with somewhat

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[1]) Anatomically this sketch is not quite exact as each scale (as is well-known) lies concealed in a pocket of the skin, from which only the posterior part emerges. Moreover these posterior parts, which together form the surface of the body, are covered by a continuous layer of pigment-bearing mucous epiderm-cells.

[2]) As everybody, who is accustomed to manipulate fresh fish knows, scales easily get loose from the surface and would no doubt do so still more quickly, if their surface was smooth on both sides.

greater energy and this shows itself on the scale as a broader ring all round a narrow, and sometimes very narrow, ring which represents the growth before the winter-rest sets in;

2nd. because the growth in the sea is a more rapid one than in fresh water; whence it is possible to distinguish rather easily on the scale of a fish returning from the sea: which is the part formed during the stay in freshwater and which is the part of the scale formed in the sea; and

3rd. because the growth, which took place in the sea in winter, can also be traced on the scale; in the sea the growth does not quite cease during that period, as is the case in freshwater; but it is slower there than in the other months and the rings then formed on the scale are in consequence narrower and separated from one another by nearly parallel ridges, forming together a group which can easily be distinguished.

These rules to some extent might already have been deduced from the results of the investigations of my predecessors: I based on them my own researches, which I think proved useful especially for the determination of the age of the salmon going to the sea and also of those returning from it for the first time (the youngest of which are the so-called "grilse"). I limited my investigations to the salmon of the Rhine [1]). The results arrived at may be distributed over the following headings: *a* and *b* young salmon, one or more years old which have not yet left the fresh water; *c* young salmon (smolts) caught in the estuary, about to begin their life in the sea; and *d* salmon, which after having spent a certain number of months in the sea, begin to ascend the river.

a. Young salmon in the first months of their life, which they spend in freshwater.

The young salmon of the upper waters of the Rhine close the larval part of their life in the course of April — partly, however, as early as the beginning of that month (sometimes even at the end of March) and partly in the beginning of May only. They are no more larvae now but small fish; their yolksack is empty, and

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[1]) My results as a whole confirm the conclusions arrived at by JOHNSTON (l. c.) with regard to the growth of the scale of the salmon. However, as there seems to exist a remarkable, hitherto unexplained difference in the habits of the Scotch salmon and that of the Rhine (a difference which lately I pointed out in an article in the "Mededeelingen over Visscherij", 1909, p. 123) with full acknowledgment of the value of the work of my predecessor, I think better for the moment not to enter into a comparison of Mr. JOHNSTON's results and mine.

so they are bound to search for the necessary food in their surroundings. The little salmon in that stage has a length of from $2^1/_2$—3 cm. and begins to develop scales on the surface of its body. From a number of observations made with the aid of fish growing up in wild condition, and from such reared in hatcheries as well, we know, that the rate of growth under the same conditions in several specimens can be very different: at the end of September the parrs in the brooks which then are 6 months old may have a length of 6, 8 or 10 cm., those reared in a hatchery-pond at the same moment may vary in length from 5 to 9 cm. Their growth continues in autumn and seems quite to cease in winter, at least during a few weeks or in some years even months. When in early spring the animal and plant life in the brooks awakes, the young salmon soon begin to feed again and to continue their growth. Young salmon of a length of from 13 to 16 cm. observed in the brooks in May may be considered as fish of 13 to 14 months old, those of 18 to 20 cm. or a little longer are over two years old.

Now let us control that growth with the aid of the scales. As questions of morphology or histology in my researches were of no concern, I began with investigating fish of a length of about 5 cm., the scales of which can easily be made out with the naked eye. For the study of the development of the scales in the first year it is an advantage, that the growth of the young salmon living in the brooks can be controlled, month after month, with the aid of fish reared in ponds; whence with regard to the growth in that period we have mathematical certainty, and this is in any case a good starting point.

The figures 1—10 refer to young salmon in the first year. The scale of the smallest fish investigated, viz. one of 4.8 cm., is figured in fig. 2; a little larger was the fish (5.5 cm.), the scale of which is represented in fig. 1 and 1a. Its shape is oval; the surface shows half a dozen concentric lines, which in the anterior part of the scale (in the figure the underpart) run close to one another and are more widely separated towards the other (exposed) extremity of the scale; five of these rings are closed, but the sixth is not: it is a regular occurrence in the scales of salmon, that the 5 or 6 innermost only, or only 5 or 6 of a certain number of innermost lines, are closed rings and that all the lines outside these remain open. [1])

The scale grows with the growth of the fish and the number of rings on the scale increases at the same rate. The surface seems not

[1]) This is different in the scales of brook-trout and sea-trout. A few remarks on the scales of these fishes will be found at the end of this paper.

to grow equally fast at all parts of the body, and I feel inclined to consider this as the reason, why for example the scales closely behind the head (above the pectoral fin, a place which is called the shoulder by some authors) remain smaller than those of the sides. The figures 3, 4 and 5 represent three scales of the same salmon, but from different places of the body and of a different size. In accordance herewith the number of rings is also different. To facilitate the comparison, all the other figures of the Plates, therefore, were drawn from scales taken from the same place on the body surface, and as such was chosen the place where a line, running from the beginning of the dorsal fin to the anterior point of attachment of the pelvic fin, would cross a line running parallel to the lateral line about three rows of scales above it.

Fishes of the same age but of different size have scales of different size and different numbers of lines of growth in consequence: the figures 6, 7 and 4 were drawn from scales of three salmon reared from the same lot of eggs and which on the same date measured respectively 5.1, 6.6 and 9.1 cm.; the numbers of their lines of growth are about 8, 12 and 16.

The figures 8, 9 and 10 show — for completeness' sake — that the growth of the scales, and the increasing of the number of lines on them, continues regularly in autumn with the growth of the fishes. So I counted:

on the scale of a parr of	7 cm.	in August	,	15	lines of growth
" " " " " " "	9.5 "	" September,		19	" " "
" " " " " " "	11 "	" October	,	22	" " "
" " " [1]) " " "	11.5 "	" November,		24	" " "

Scales of salmon caught in December are the last of fishes of the first year, which could be investigated: it becomes difficult already in the later autumn months to catch young salmon, and it is quite impossible to do so as soon as the brooks in which the parr pass the winter, are frozen. As soon as these brooks thaw again in early spring and swell with the snow melts on hills and mountains the young salmon return to activity and most of them profit by the swelling of the brooks and the descent of the water to find their way to the main river and thence to the sea. We will return to these in *c*, page 493. Some of the young salmon, and perhaps not so few, however, remain at the end of their first year in the brooks of the upper course, most probably at or near the places where they were spawned. These are:

[1]) This scale is not figured.

b. Young salmon, which at the end of their first year do not leave the upper course of the river.

From April or May, in numerous brooks of the upper course, it is not so difficult to catch young salmon of a length of 13 cm. and more. It is not possible that these were born in the same year and they must be over one year old in consequence. How it is, that part of the young salmon remain behind on the upper course, when the great majority set out for the main river and the sea in early spring, is not sufficiently explained. We only know, that sex is concerned in the matter: a very large percentage of the remainder, one year old fish, are males and the still older ones are, as far I could investigate, all males and all these males become mature and spawn before going to the sea. It is obvious, therefore, that the remaining behind is connected with the early development of maturity in the males; yet the question is a somewhat complicated one 1. because other males (and most probably the greater part) descend to the sea along with the females at the end of the first year; and 2. because some females also remain in the river at the end of the first year.

The unequal growth, which in the salmon of the first year is so striking, seems to continue to exercise its influence; to control the age, therefore, the growth alone is of little use: the structure of the scales renders excellent services in the matter. In May you can catch small salmon in the upper course of the Rhine from 13 to 20 cm. long; in July as long as 24 cm.; in August of the same size, but also some of 13, 14 etc. cm.; in September larger ones of 20 and more cm. but still some of from 15 to 17 cm. and even in October and November the smallest salmon caught, were not longer than 14 cm.

The figures 11—13 were drawn from scales of such salmon: figures 11 and 12 are both from those of salmon caught in August. Both show the formation of the first year and around it what has been formed since the beginning of the second year. Whereas the one salmon (fig. 11) in the first year developed only insufficiently — as may be concluded from the small dimension and the small number of lines of the first year's scale — the scale of fig. 12 is of a much better grown fish. Comparing fig. 13 with the foregoing ones, little doubt remains as to the age of this salmon: it is a parr of a length of over 20 cm. caught in the Kinzig in May, which spent two whole years in the upper course of the river, and which, on being caught, was in the beginning of its third year. The limits of the parts formed in the one and the next year in these fishes present themselves as a somewhat more prominent line; the rings

formed in the first year are, moreover, very narrow, those of the second year are wider; finally, those formed in the beginning of that second year can often be distinguished from the rings formed later in the same year by a somewhat greater width.

A part, most probably a very great part, of the two year old salmon, with the majority of the one year old parrs, leave the upper course of the Rhine in early spring to reach (in May) the mouth of the river in Holland and thence the salt water. There can be no question that but a few — like the little fish, the scale of which is seen in fig. 13 — remain in the river over two years. It is generally admitted, however, that these will also find their way eventually to the sea.

c. The young salmon, which are about to enter the sea.

Some years ago (1896) I had an opportunity of investigating between three and four hundred young salmon (smolts), which were caught in May in one of our estuaries and which were on the point of arriving at the sea. Regarding the age of these salmon I was only able to say [1]), that the curve drawn for the length of these salmon seemed to demonstrate, that they were all of the same age. The length of these salmon varied from 11.5—21.3 cm., no less than 95 % of them, however, were from 12—17.5 cm. A sample was preserved in spirits and so it was possible (in 1909) to investigate forty specimens as to their age with the aid of their scales. Their length was from 12—17.5 cm. Of these 26, length varying from 12—16 cm., were upwards of one year old; and 13, length from 14.5—17.5 cm., were upwards of two years old. That the length alone is a bad criterion for the determination of the age is shown by the circumstance, that of 19 of these smolts, of from 14.5—16 cm., 12 were found to be upwards of one year and no less than 7 upwards of two year old fish.

The figures 14 and 15 are from the scales of two of these fish. They leave no doubt as to their age: the first is from the scale of a smolt of 12.6 cm., the age of which, in May, was about 14 months, the other from a smolt of 16.8 cm., which at the moment of the catch was about 26 months old. The first shows the margin, which indicates the period of rest in the first winter, and also the not very numerous rings of growth formed since that period; in the other specimen the lines indicating the period of rest in both winters are easily distinguishable.

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[1]) Hoek, Neuere Lachs- und Maifischstudien. Tijdschr. d. Ned. Dierk. Vereen. (2). VI. 1899.

The figure 16 represents the scale of a smolt of 21.3 cm., the only fish of that length caught with the smaller ones in the mouth of the Hollandsch Diep. At first I took that fish for a salmon over three years old, later on, however, I changed my opinion: I now think that it is a fish also about 26 months old, but one that is somewhat better developed. We now know, that there are young salmon which remain in the brooks more than two years; though it is probable, that these as fully three year old fish would descend to the sea along with those of one and two years, yet this has never been observed with certainty. In the upper regions of the river young salmon have been observed up to a length of 24 cm., such long ones, however, have not been seen there after August or September. Should this rule hold good, it would prove that an autumn-migration takes place consisting perhaps of larger and not very numerous specimens which, from being more isolated swimmers, are not so easily discovered as those descending in denser shoals in spring [1]).

d. Salmon returning from the sea.

The smallest of the returning salmon (the grilse or Jacob-salmon as they are called in Holland) make their appearance in the river slightly earlier in one year than in another. As a rule the first are caught at the end of June, they are most numerous, however, in July and August among the salmon then ascending the river. Their length varies from 55– 67 cm., those of 55—60 cm. are often called "small" grilse and those of 64—67 cm. large ones.

The investigation of the scales of two small grilse of 55.6 and 57 cm. brought to light, that these fish, which were caught in the lower part of the Rhine on August 9th and 14th respect., only passed one winter in saltwater. A winter passed in the sea is shown on the scale in a somewhat other manner than is the case in freshwater. As has been pointed out already, the growth of the body and, in correspondence herewith, that of the scale ceases absolutely in the frozen brooks; when the growth begins again in early spring, it marks itself out on the border of the scale as a somewhat broader ring, (usually an extremely narrow ring first and then that broader one): so each winter passed in freshwater is easily recognised. In the sea the salmon go to places where the water is deeper and here the

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[2]) Years ago, two young salmon of 12.5 and 14 cm. were sent to me from Gorkum on the Merwede, which were caught in a lamprey-pot, in November 1886. Other instances of young salmon caught in autumn at or near the mouth of the Rhine in Holland have not come to my knowledge.

fall in temperature in winter is by no means so considerable as in the river: there is never any question of the sea freezing and an absolute cessation of the growth in consequence. Yet even there down to the bottom and also in deeper water the vital processes in winter are more or less dependent on the surface, which is more directly under the influence of sunshine or of atmospherical phenomena in general. Now that lesser activity of all the vital processes can be traced on the surface of the scale in the shape of a sometimes narrower, sometimes broader group of narrow rings of growth between the others developed in the sea, and which as a rule are much broader. The part of the scale formed in the sea is recognised at first sight: here the lines of growth run at a greater distance from one another; they are more loose and irregular when compared with the nearly smooth and parallel lines on the part formed in freshwater. The marine part, moreover, is as a rule formed more quickly than the freshwater-part, which is quite in accordance with the more rapid development of the whole body. We saw that the length a young salmon has at the end of the first year is 16 cm. at most and that, in freshwater, as a rule they do not reach 20 cm. in their second year. If, however, such a young fish of 16 cm. at most descends to the sea in May of a certain year, it may come back next year in July or August as a grilse of a length of 55—57 cm. The figures 17 and 17a were drawn from a scale of such a grilse. There is no doubt in my mind, that we have to do with a young fish hatched in the spring of 1907, which had a good development during its first year and which found its way to the sea in May 1908. Its length at that time was 16 cm. at most and its weight may have been 55—60 grams. In the sea it grew rapidly during a period of one year and three months and in August of the following year it came back from the sea as a fish of a length of 55 cm. and of a weight of 1.44 kilogrammes. [1])

Only part and a relatively small part of the smolts which have migrated to the sea, return so soon. Part already of those returning as grilse stayed a second winter in salt water before beginning their ascent of the river. Of such an older grilse (of a length of 64 cm.) a

[1]) In the natural history of the salmon this is a point which has always given rise to great controversy and on which even now no general agreement exists. I may remind the reader of the famous "Stormontfield experiments" so often cited in the literature of the subject, which (1854—61) were taken with marked fishes and which should have proved that smolts of half an ounce weight, which went to the sea in May, returned six weeks later to the river as grilse of a weight of $3^1/_4$ pounds. I think we may say now, that even in the sea salmon do not grow with such rapidity.

scale is figured in fig. 18 and 18a. There can be no doubt but that this fish spent two winters in the sea before being caught. On the other hand I am by no means so sure with regard to the earlier data of this fish. The scale of the grilse of fig. 17, roundabout the oldest part, i. e. the one formed during its stay in freshwater, shows only a few lines of growth, developed in the first weeks or months of the second year, which this fish, before it went to the sea in May, still spent in the river and to these rings the parts formed in the sea closely joined. In the scale of fig. 18 the part roundabout the first year's scale, which part probably is also formed in freshwater, is much broader — but, as is the case in the scale of fig. 17, it goes over without any sharp limit into the marine growth of the scale. If I had not such good reasons for believing that on the Rhine the migration of the smolts to the sea generally takes place in May, and if what I have seen of migration in other months (e. g. in autumn) was not so extremely scanty, I should be inclined to suggest, that this salmon spent a second summer in freshwater, went to the sea in autumn, and thence returned to the river, after having passed two winters in the sea. This grilse was 3 years and about 5 months old on being caught in August, and of that period $2^1/_4$, or, if the second supposition is correct, $1^3/_4$ years were spent in the sea.

One need not wonder that such cases of uncertainty as the one mentioned above still occur. The method of using the scales to determine the age of the salmon is comparatively new. Yet I think that my communication, being based partly on observations of the fish and partly on the study of its scales, may also contribute to give that method a sound basis. Therefore, though my researches are far from finished, I thought it useful not to delay the publication of these still provisional results. This question with regard to the salmon is a complicated one: we have to reckon with the possibility of the parrs staying one or more years in freshwater, before they migrate to the sea; with the exodus in May or perhaps in another month of the year; with their return from the sea in different seasons, and in a very varying stage of sexual development. For the older and larger fishes moreover a difficulty arises from the circumstance, that in any case some salmon (and perhaps not so few) after having spawned, swim down the river again, later to ascend it for the second time. These fish, which are called "kelts", seem to return in greater quantities in April and May; in Holland at least a certain number of them, on their way to the sea, are regularly caught about that time: in this year over 100 of these fishes were taken. Now, when these fish will have reached their feeding grounds in the sea again, 6 or 9 or 12

months may have elapsed, since their departure and since they ceased to take food, — it is even possible that they spent a considerably longer period still, without taking food in the normal way, which is done by the older salmon only in the sea, as is well known. Hence it is possible that the growth of the fish itself and of its scales has also been suspended for so long a time. If we wish to consider the length of the periods and the regularity in the alternation of the stay in freshwater and salt, we have to reckon with all these possibilities; hence one cannot be surprised if the picture of the growth, which the scale presents. is often not easy to understand. Personally as yet I have no great experience in the matter[1]) as I could not include in my investigations either the scales of kelts or those of very big and old salmon which, probably, do not ascend the Rhine for the first time. I investigated, however, a single, so-called small, and a single, so-called large, summer salmon and I shall give here a summary description of their scales.

The figures 19 and 19*a* are from the scale of a small summer-salmon of 84 cm., which was caught in August at one of our seine-fisheries. The scale shows the formation of the first year in rather good development. The number of rings formed in that first year is 16—18, if we count them in oblique direction from the centre to the rounded corner where the lateral and front (in the drawing the under) margins meet. The section grown in the second year consists of two parts: first (going from the interior to the exterior) 14—15 rather regular rings, which are slightly broader than those of the first year and which represent, with the latter, the growth in fresh water; then 9—11 somewhat less regular rings, the most interior of which are considerably broader: they are the first rings formed in the sea. Roundabout that division a few very narrow rings are to be seen, which represent the growth of the winter at the end of the second year. In the third year about 15 nearly equally broad rings have been formed and roundabout these a group of 3—4 narrow rings, with here and there perfectly parellel margins, which seem to indicate the growth of the winter of that year. In the same way the growth of the fourth year is formed by 25—27 such rings and the growth of the third winter passed in the sea by a band of from 4 to 5 narrower rings with nearly parallel margins. Finally that part of the 5th year, which lasts from winter until August, is represented

[1]) JOHNSTON (l. c.) describes the scales of such salmon; what he tells us about the "spawning marks" which are to be seen on the scales of older salmon is very interesting and in any case is worth being compared with what the scales of Rhine-salmon will show under the same circumstances.

by a dozen nearly equally broad rings. Therefore, in my opinion, this small summer salmon, which was caught in August 1909, was probably one of the winter 1904/1905 spawn.

Quite in accordance with what I expected I then found that a so-called large summer-salmon (of a length of 1.024 m.), caught on the same day as the smaller one just described, and the scale of which is represented by the figures 20 and 20*a*, was still one year older to judge from the structure of that scale. The analysis of the scale is the following:

A rather large central division with 23—25 rings represents the first year.

Roundabout this part the formation of the second year is seen: it consists of 6—7 rings formed during the stay in freshwater and 12—14 formed in the sea and it is separated from the partition formed in the third year by an indistinct [1]) and narrow ring.

The part of the third year is composed of about 14 broader rings and is surrounded by a group of narrow rings, which probably represent those formed in winter time, the breadth of this group only slightly increasing towards the middle of the scale.

Next comes a slightly broader band built up of 17—18 rings. This band seems to form the growth of the fourth year and is enclosed by a not very prominent winter-band, which consists of five narrow rings in the middle, and which greatly diminishes in breadth on both sides.

Upon these we have a much narrower band, which grows extremely narrow, in the middle of the scale, owing to the winterband (which separates it from the division formed in the last year of this salmon's life) being very broad there. This winter band in the middle is composed of about 10 narrow rings, the margins of which are very distinct and nearly quite parallel to each other.

Finally we have roundabout that division the part which represents the growth of the 6th year, which was broken off in August 1909 and which shows itself as a band of 16—18 first broader, later on narrower rings.

According to this sketch the ring formed in the fifth year would be remarkably narrow. Have we perhaps to do with one of the fishes, which visit the river for the second time? And is the not very distinct winter-band between the growth of the 4th and 5th year together with the narrow section of the scale formed in the 5th year

---

[1]) In the part figured (fig. 20) this separation can only be distinguished owing to the first ring of the next year being very broad. In fig. 20a it is indicated as a single line.

to be considered in connection with a winter period passed on the upper course of the river? It is impossible to answer these questions with the material at present at my disposal.

I think my description shows that, whereas the application of the method has already given some interesting results with regard to the growth and the age of the Rhine-salmon in the first years of their life, for the age and the growth of older fish several obscure points are still waiting for elucidation [1]). To begin with it will be very important to study the scales of the kelts descending to the sea in early spring and especially, to compare their structure with those of the so-called winter-salmon, the name we are accustomed to give to big clean fish, the largest of all ascending the river. As I have already said for that investigation no material was at my disposal, but I intend to continue these researches and will with pleasure report again on them later. Here a few more remarks may find a place regarding the scales, drawings of which are reproduced in the figures 21—23.

Fig. 21 was drawn from the scale of a salmon of a length of 20.4 cm. and shows a somewhat abnormal central part. Such scales are by no means rare; they have instead of the usual centre beginning as a very minute ring and narrow rings encircling it, a considerably larger irregularly defined central part of an oval or more rounded shape. Sometimes several of these scales are observed in close proximity, normal ones lying between the abnormal scales. It is obvious, I think, that these abnormal scales are such, the development of which was retarded, so that a centre of much greater dimension was to be formed at once. It is also possible that they occupy, like so many new formations, the places of other scales which the fish lost by accident. The earlier investigators of fish-scales (STUART THOMSON and JOHNSTON) also mention these abnormal scales but neither of them pronounce themselves in a decided way on their origin.

Figure 22 represents the scale of a brooktrout (*Salmo fario*) and figure 23 one of a seatrout (*Salmo trutta*). To distinguish smaller specimens of these fishes from young salmon (*Salmo salar*) of about the same size is even now a somewhat difficult affair. Therefore I think it is important, that the structure of the scale in the first

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[1]) By saying so I do not mean to be prejudicial to the work of my Scotch predecessor (Mr. JOHNSTON). The salmon of the Tay formed the main object of his investigations and he pointed out himself, that it was not probable that the statistical particulars of Tay scales would apply even to those of all other rivers (of Scotland) without any modification.

years, regularly affords a good mark to distinguish young salmon and trout as far as I could ascertain. In the scale of thé salmon 5 to 6 and no more of the very first formed rings are quite closed: their shape is oval and they are surrounded by the larger ones all of which have the shape of U's. With both species of trout it seems to be the rule, however, that of the rings formed on the scale in the first years a much larger number present themselves as quite closed rings. In the seatrout (fig. 23) this is the case with the rings formed in the first year especially, in the brooktrout (fig. 22) a very large proportion of the rings of the second year show this particularity: sometimes with a few exceptions all the rings of these years are so formed. So the fact that there is a greater relationship between brooktrout and seatrout (called Scotch salmon by our fishermen) than there is between the salmon and one of these trout, which was pointed out by DAY[1]) years ago and which I emphasized myself on a former occasion [2]), by the structure of the scales is confirmed in a very striking way.

EXPLICATION OF THE FIGURES.

N.B. All the figures are drawn with the camera, so there can be no question regarding the exactness of the shape and the main lines; the lines of growth, however, were free hand drawn, their number as much as possible in correspondence with the object. All figures are drawn with the free end of the scale above. This end is directed backwards on the fish.

PLATE I

Fig. 1–13: Scales of salmon, *which have not left the freshwater as yet.*

Fig. 1—10: Salmon *of the first year.*

Fig. 1. Scale from the side of a little salmon of 5.5 cm., caught Aug. 12th, 1909. Magnified 148 diameters.

Fig. 1*a*. Same scale, magnified 30 diameters.

Fig. 2. Scale from the side of a little salmon of 4.8 cm., caught end June 1909 in the so-called Brook of Hüningen. Magnified 30 diameters.

Fig. 3, 4, 5. Scales of a little salmon of 9.1 cm. from Marxzell, September 28th, 1909, from different places of the surface of the body. Magnified 40 diameters.

Fig. 3 taken a little behind the head.

„ 4 „ from the side, above the lateral line.

„ 5 „ about the height of the anal aperture, beneath the lateral line.

Fig. 6. Scale from the side of a little salmon of 5.1 cm. from Marxzell, September 28th, 1909. Magnified 40 diameters.

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[1]) FR. DAY, The Fishes of Great Britain and Ireland. 1880—84.

[2]) HOEK, La truite de mer du Rhin. Tijdschr. d. Ned. Dierk. Vereen. (2). III. 1892. p. 243.

Fig. 7. Scale from the side of a little salmon of 6.6 cm. from Marxzell, September 28th, 1909. Magnified 40 diameters.
Fig. 8. Scale from the side of a little salmon of 7 cm. from the Gaybach, August 13th, 1909. Magnified 30 diameters.
Fig. 9. Scale from the side of a little salmon of 9.5 cm. from the Prüm, September 1909. Magnified 30 diameters.
Fig. 10. Scale from the side of a little salmon of 11 cm., caught in October 1909 in the Rhine near Basel. Magnified 30 diameters.

Fig. 11—13: Salmon *more than one year old.*

Fig. 11. Scale from the side of a little salmon of 13 cm., caught in the Kinzig August 16th, 1909. Its age is about 17 months; it has not grown so well in the first year. Magnified 30 diameters.
Fig. 12. Scale from the side of a little salmon of 15.1 cm., caught in the Kinzig August 16th, 1909. It is of the same age as the foregoing but it has grown well in the first year. Magnified 30 diameters.
Fig. 13. Scale from the side of a little salmon of 20.5 cm., caught in the Kinzig May 13th, 1909. It has entered on its third year. Magnified 30 diameters.

Fig. 14—16: Scales of salmon caught in the mouth of the Hollandsch Diep, shortly before they would have arrived in the sea.

Fig. 14. Scale from the side of a little salmon of 12.6 cm., caught in May 1896, at the age of about 14 months. Magnified 30 diameters.
Fig. 15. Scale from the side of a little salmon of 16.8 cm., caught in May 1896, at the age of about 26 months. Magnified 30 diameters.
Fig. 16. Scale from the side of a little salmon (perhaps a seatrout) of 21.3 cm., caught in May 1896, most probably of the same age as the one the scale of which is figured fig. 15. Magnified 30 diameters.

Fig. 17—20: Scales of salmon which, *after having passed one or more years in the sea,* have returned to the river.

Salmon from the market at Kralingsche Veer.

Fig. 17. Part of the scale from the side of a grilse of 55.6 cm.; caught August 14th, 1909. Magnified 30 diameters. This fish which spent over a year in the fresh water, stayed in the sea from May 1908 — August 1909: about 15 months.
Fig. 17*a*. The same scale entirely. Magnified 4.2 diameters.
Fig. 18. Part of the scale from the side of a grilse of 64 cm., caught August 6th, 1909. Magnified 30 diameters. This fish after having spent over a year (perhaps about one year and a half) on the river, stayed in the sea from May 1907 (perhaps from the autumn of that year) to August 1909: about 27 (perhaps 22) months.
Fig. 18*a*. The same scale entirely. Magnified 4.2 diameters.

PLATE II

Fig. 19. Part of a scale from the side of a so-called small summer salmon of 84 cm., caught August 6th, 1909. Magnified 30 diameters. This salmon came into the sea after having spent over a year in freshwater and returned to the river after a stay in the sea of about 39 months.

Fig. 19*a*. The same scale entirely. Magnified 4.2 diameters.

Fig. 20. Part of a scale from the side of a so-called large summer salmon of 102 cm., caught August 6th, 1909. Magnified 30 diameters. This salmon after having spent over a year in the freshwater descended to the sea and stayed there for about 51 months, perhaps broken by a visit to the river.

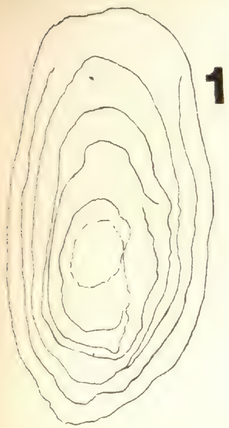
Fig. 20*a*. The same scale entirely. Magnified 4.2 diameters.

Fig. 21. Scale from the side of a young salmon of 20.4 cm., caught in the Kinzig May 13th, 1909. Magnified 30 diameters. In this scale the formation of rings in the first months seems to have failed — probably the scale is a new product formed in the place of a scale which was lost.

Fig. 22. Scale from the side of a brook-trout (*Salmo fario*) of 17 cm., caught in the Hohlbach November 1909. Magnified 30 diameters.

Fig. 23. Scale from the side of a sea-trout (*Salmo trutta*) of about 45 cm., caught in the Baltic near Bornholm. Magnified 30 diameters.

(January 12, 1910).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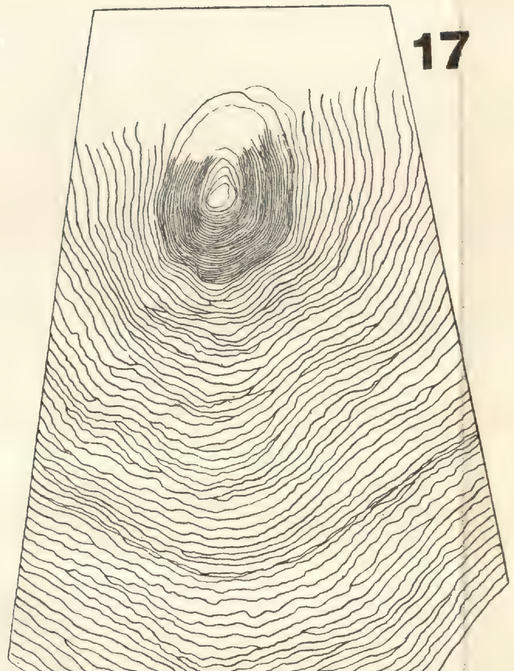
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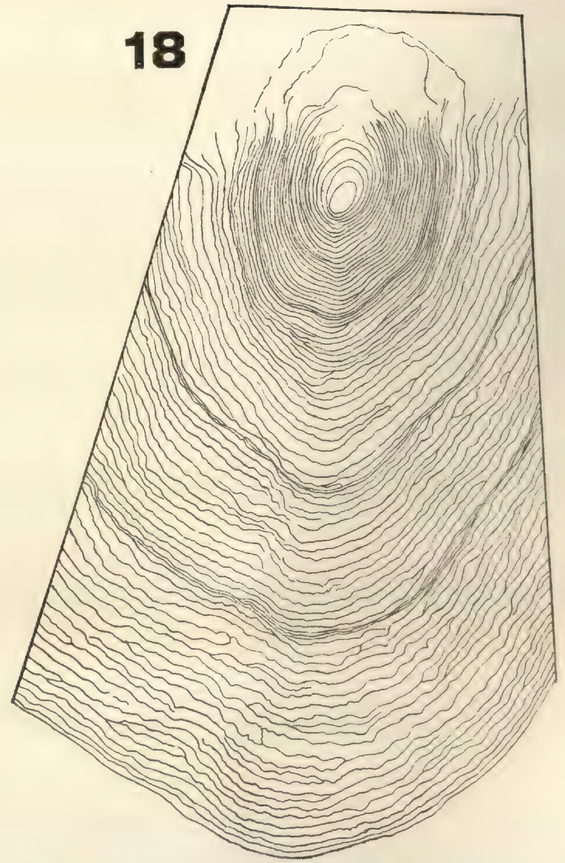
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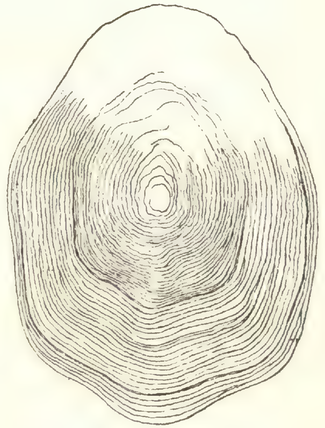
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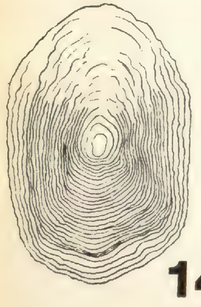
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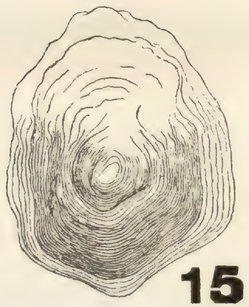
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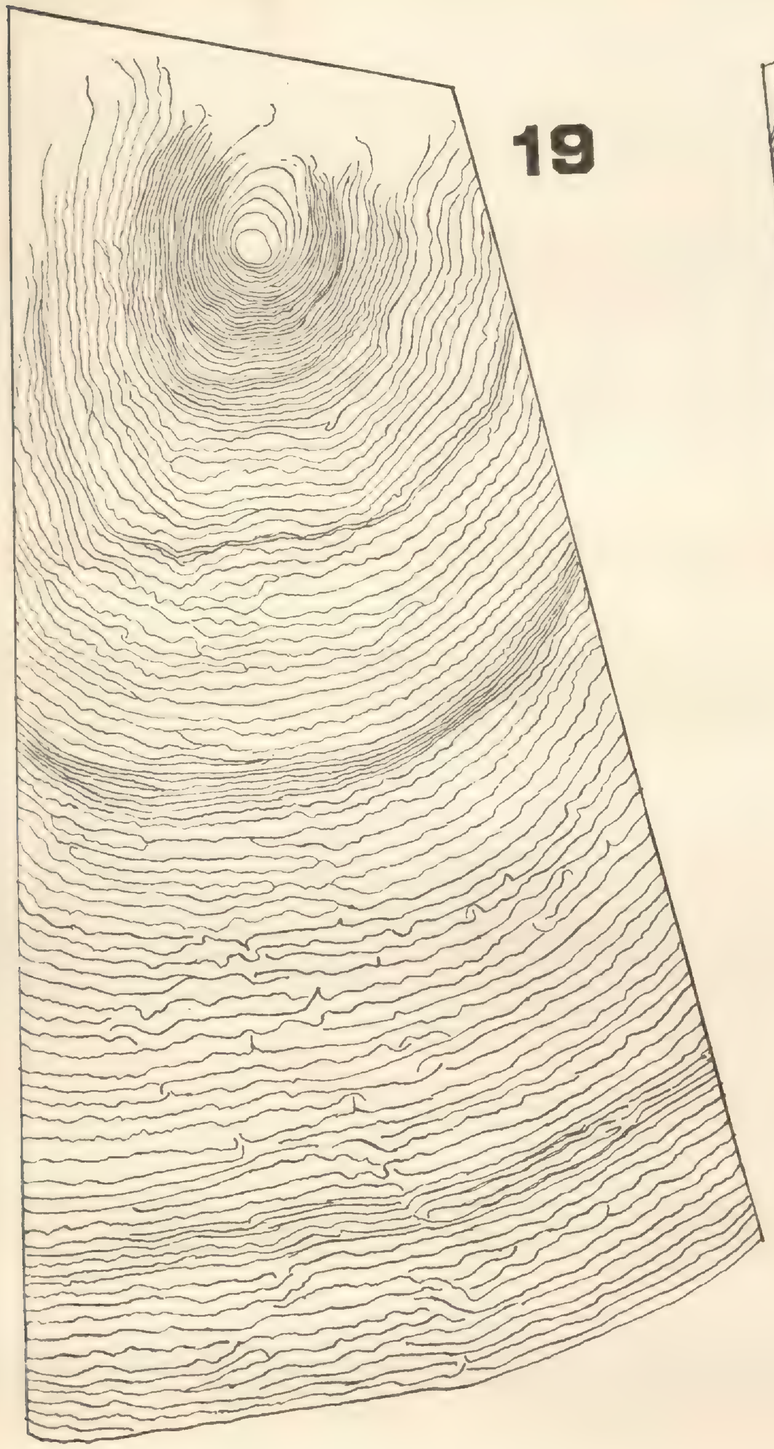
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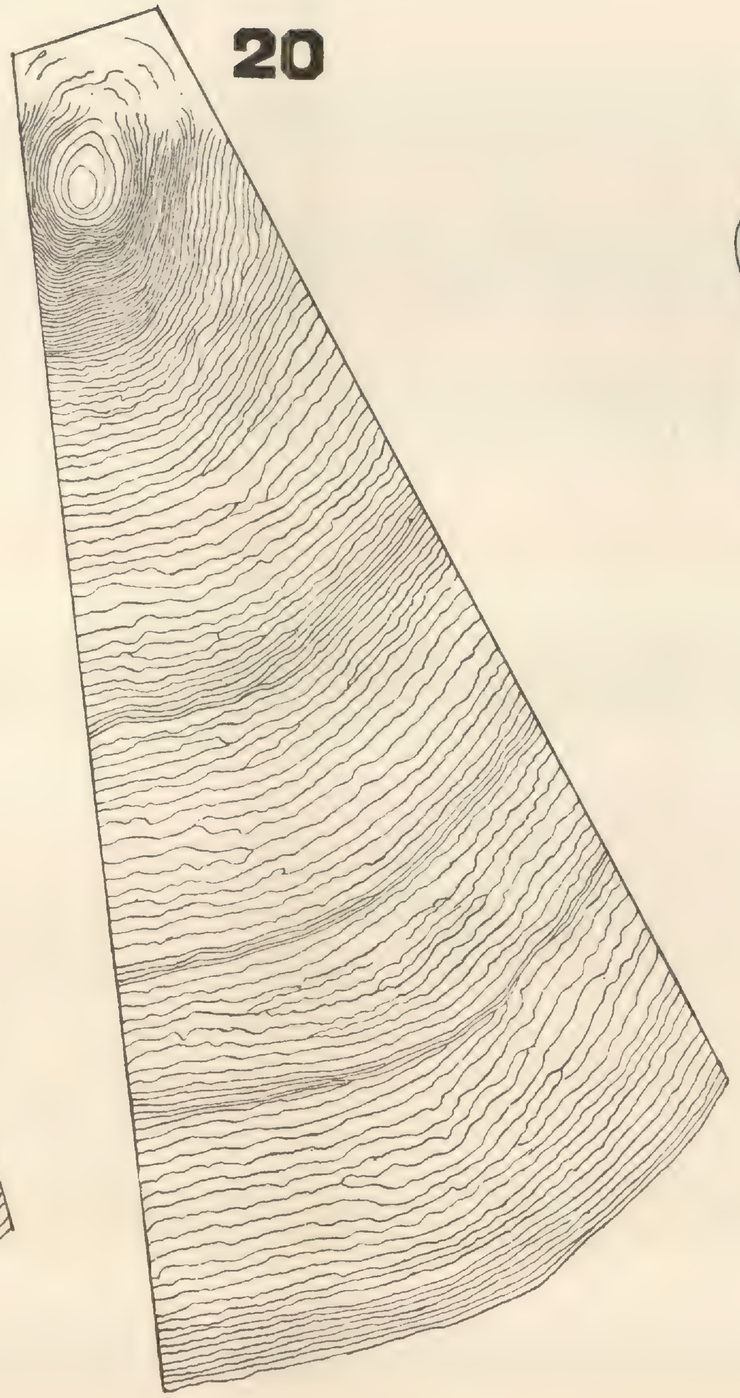
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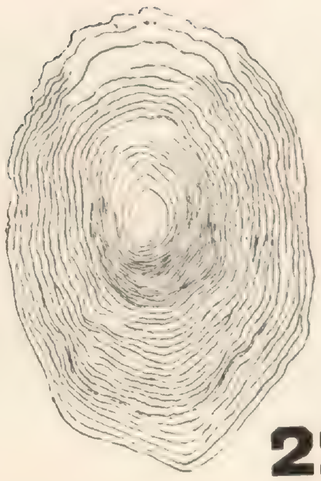
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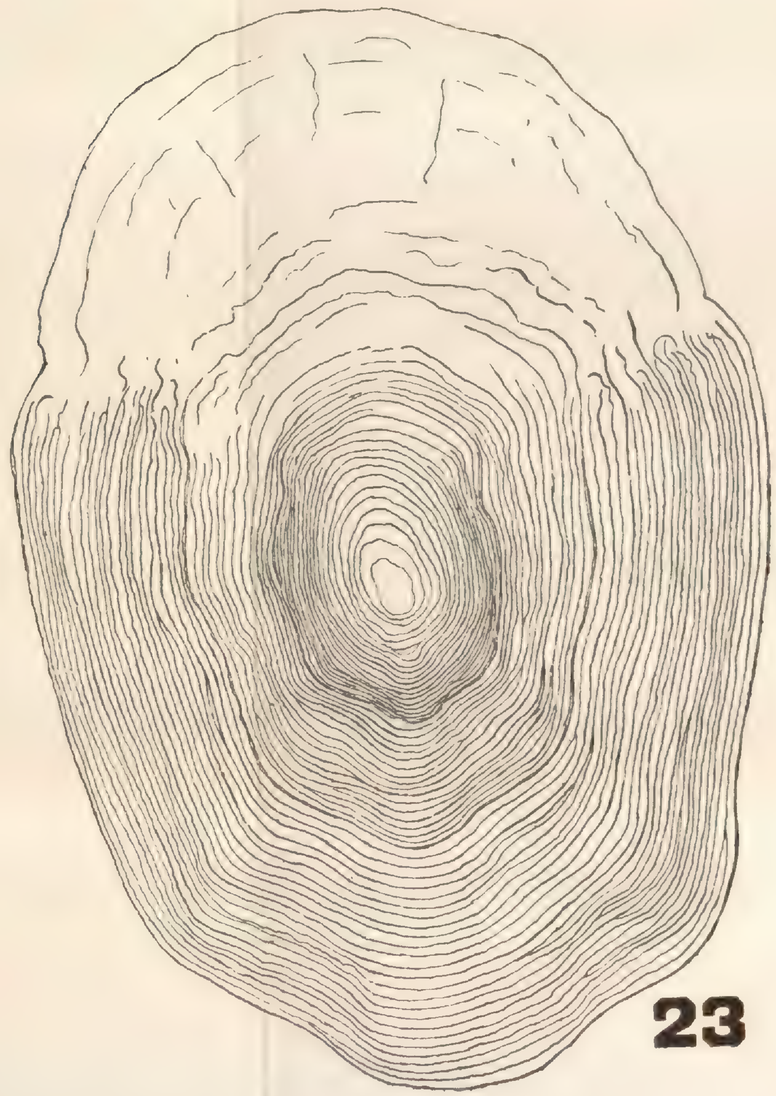
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PROCEEDINGS OF THE SECTION OF SCIENCES

VOLUME XII
(— 2ND PART —)

JOHANNES MÜLLER :—: AMSTERDAM
:———: JULY 1910 :———:

(Translated from: Verslagen van de Gewone Vergaderingen der Wis- en Natuurkundige Afdeeling van 24 December 1909 tot 29 April 1910. Dl. XVIII.)

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KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN
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PROCEEDINGS OF THE MEETING

of Friday December 24, 1909.

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Mathematics. — "*On the orbits of a function obtained by infinitesimal iteration in its complex plane.*" By M. J. VAN UVEN. (Communicated by Prof. W. KAPTEYN).

(Communicated in the meeting of November 27, 1909).

When a function $y = \varphi(x)$ is iterated, each iteration $y_n = \varphi_n(x)$ will give rise to a conform representation of the complex planes of x and y_n.

If we suppose $y = \varphi(z)$ to be built up by means of infinitesimal iteration of the function $\underset{m=\infty}{Lim}\, y_{\frac{1}{m}} = \underset{m=\infty}{Lim}\, \varphi_{\frac{1}{m}}(x)$, so that y_n has also a meaning for broken and unmeasurable values of n, then the conform representation of $y = \varphi(x)$ will gradually appear out of the identity belonging to $y_0 = \varphi_0(x) = x$.

We now regard a plane V_0 as complex plane of the quantity x and we place the complex plane V_1 of the quantity $y = \varphi(x)$ parallel to V_0 at a distance h and in such a way, that the real axes and the imaginary axes are each other's orthogonal projection. Then to each point x of V_0 are conjugated by means of the function $y = \varphi(x)$ one or more points y of V_1. By connecting corresponding points x and y by rays a congruence of rays is formed which can serve as the image of the function $y = \varphi(x)$.

For the case $y = \varphi(x) \equiv x$ we should obtain in this way the congruence of rays formed by all the normals on the planes V_0 and V_1 as representative of the identity.

If now we let the function $y = \varphi(x)$ gradually arise from the identity, then to each stage of the generating process a definite congruence of rays will belong. All these congruences form together a complex of rays. It is clear, that the formation of the function $y = \varphi(x)$ will now be represented by this complex of rays.

Let us first examine the complex cones of the points of V_0. Each point $x = u + iv$ of this plane is the vertex of a cone counting in any case the normal in x on V_0 among its generatrices; this edge namely intersects the plane V_1 in $y = u + iv = x$.

The section of this complex cone with V_1 will pass through the point $z = x$ and all points representing the values taken by $y_n = \varphi_n(x)$ when n increases from 0 to 1. So this section also gives us a representation of the generating process of $\varphi(x)$. It goes without saying that we can continue the iteration also past $y = \varphi(x)$ and likewise that we can also regard negative values of n. The whole of the complex cone embraces in fact all functions $y_n = \varphi_n(x)$, where n varies from $-\infty$ to $+\infty$. Also the section regarded as a whole will contain all the values of the function $y_n = \varphi_n(x)$, where x is constant and n varies from $-_\infty$ to $+\infty$. Each value of x possesses its own complex cone and therefore also its own section. We shall indicate this section by the orbit $x \rightarrow y_n$.

We might also have indicated the increase of $\varphi(x)$ by allowing the plane V_1 to grow gradually out of V_0 and that by allowing the distance of the planes to increase regularly from 0 to h, so that $\varphi_n(x)$ is represented in the plane V_n at a height nh above V_0. Let us then suppose in each plane V_n the image $y_n = \varphi_n(x)$ belonging to some initial-point $x = u + iv$ to be constructed, then all these points will form in their regular succession a twisted curve. Each of the ∞^2 points x of V_0 gives rise to a suchlike *twisted curve* and the function $y = \varphi(x)$ with its different stages of development is thus represented by a *congruence of twisted curves*.

It is clear that the orthogonal projection of the twisted curve of x on the plane V_1 coincides with the orbit $x \rightarrow y_n$.

We shall for the present occupy ourselves only with the study of such an orbit $x \rightarrow y_n$.

To find the orbit $x \rightarrow y_n$ we have but to solve the functional equation of ABEL. We have namely to find that function $f(x)$ of x increasing with n when for x is substituted $y_n = \varphi_n(x)$; this function increases for the process of iteration with real contributions, i.e. the quantity $\zeta = f(x) = U + iV$ describes in its complex plane the right line $V = c$ parallel to the real axis. If once we know the form of the function $\zeta = f(x)$, then we also know the orbit of the quantity $x = f_{-1}(\zeta)$.

The value of V and the initial value ($n = 0$) of the real part U of ζ represent together two arbitrary constants, of which we do not dispose until we choose the initial value of x.

We shall indicate the current point (y_n) of the orbit $x \rightarrow y_n$ by z, whilst we shall point out x by z_0; we then have

$$f(z) = f(z_0) + n$$

or

$$U + iV = U_0 + iV_0 + n,$$

so that

$$U = U_0 + n \quad , \quad V = V_0.$$

The choice of the initial point z_0 now determines the values U_0 and V_0.

When working out some examples we shall not always follow the systematic way sketched above, as it is unnecessarily lengthy in simple cases.

In reference to the broken linear function $y = \frac{\alpha x + \beta}{x\gamma + \delta}$ we notice that this has been thoroughly investigated already by POINCARÉ [1]) and KLEIN [2]), the latter having also included complex values of α, β, γ, and δ in the study. KLEIN too allows the function $y = \frac{\alpha x + \beta}{\gamma x + \delta}$ to arise gradually out of x and regards the orbit described thereby. For the non-parabolic cases he builds up the function by infinitesimal iteration in the sense indicated by us. For the parabolic case, on the other hand, he takes as parameter of the function in its orbit not the iteration-index n, but a complex multiple of it. In consequence of

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[1]) POINCARÉ. Acta Mathematica I (1882), p. 1.

[2]) KLEIN—FRICKE. Vorl. ü. d. Theorie der ell. Modulfunktionen (TEUBNER, 1890), p. 165.

this the orbit of z found by KLEIN differs a little from ours. Although after stating and annulling this difference we might suffice with a reference to the results of KLEIN, we will dwell a little longer on the function $y = \frac{\alpha x + \beta}{\gamma x + \delta}$, the more so as, differing from KLEIN, who treats first simple cases and then applies the principle of transformation of the circle correspondence, we shall immediately investigate the most general case.

Examples:

I. $y = x + \beta, \quad y_n = x + n\beta$ or $z = z_0 + n\beta$.

The point z describes the *right line* connecting the points $z = z_0$ and $z = z_0 + \beta$, in such a way that the distance from z to z_0 is proportional to n.

II. $y = \alpha x, \quad y_n = \alpha^n x$ or $z = \alpha^n z_0$.

Let us put $z = \varrho e^{i\theta}$, $z_0 = \varrho_0 e^{i\theta_0}$, $\alpha = \sigma e^{i\tau}$, then

$$\varrho e^{i\theta} = \sigma^n e^{in\tau} \varrho_0 e^{i\theta_0},$$

from which ensues

$$\varrho = \sigma^n \varrho_0, \quad \theta = \theta_0 + n\tau \quad . \quad . \quad . \quad . \quad , \quad . \quad (1)$$

or

$$\varrho = \varrho_0 \, \sigma^{\frac{\theta - \theta_0}{\tau}} = ce^{\theta}.$$

Point z describes a *logarithmic spiral* round the origin. The polar angle θ increases uniformly with n, i.o.w. the polar angle θ increases *arithmetically uniformly*; it is clear that the radius vector ϱ increases *geometrically uniformly*.

If α is real, then $\tau = 0$. The second equation (1) tells us that the polar angle remains constant, so that point z moves along the *line connecting* O and z_0 and that with a geometrically uniform increase of ϱ.

If $mod\, \alpha = 1$, then $\sigma = 1$. The first equation (1) then indicates, that the radius vector remains constant, so that point z describes the circle round O as centre, passing through point z_0. The polar angle θ increases arithmetically uniformly.

If τ is commensurable with π, i. e. if α is a root out of unity, then $y = \alpha x$ leads back to x after a whole number of iterations.

III. $y = \alpha x + \beta, \quad f(x) = \dfrac{log\left(x + \dfrac{\beta}{\alpha - 1}\right)}{log\, \alpha}$.

$$f(x) = \frac{-\infty}{log\, \alpha} \text{ for } x = -\frac{\beta}{\alpha - 1} = g, \text{ therefore } f(z) = \frac{log\,(z - g)}{log\, \alpha}.$$

If we displace the origin to g and if accordingly we call $z - g = \varrho' e^{i\vartheta'}$, we find for the orbit of z the *logarithmic spiral* $\varrho' = ce^{\gamma\vartheta'}$ round the point g. If, however, α is real, then z describes the line from z_0 to $\alpha z_0 + \beta$, containing also point $g = -\frac{\beta}{\alpha - 1}$. Is on the contrary *mod* $\alpha = 1$, then the orbit of z is a circle round g as centre.

IV. $y = \frac{\alpha x + \beta}{\gamma x + \delta}$, where $(\alpha - \delta)^2 + 4\beta\gamma \gtrless 0$.

$$f(x) = \frac{1}{\lambda} log \frac{px+1}{qx+1}, \text{ where}$$

$$\lambda = log \frac{(\alpha+\delta)+\sqrt{(\alpha-\delta)^2+4\beta\gamma}}{(\alpha+\delta)-\sqrt{(\alpha-\delta)^2+4\beta\gamma}} = log \frac{\{(\alpha+\delta)+\sqrt{(\alpha-\delta)^2+4\beta\gamma}\}^2}{4(\alpha\delta-\beta\gamma)},$$

$$p = \frac{(\alpha-\delta)+\sqrt{(\alpha-\delta)^2+4\beta\gamma}}{2\beta}, \quad q = \frac{(\alpha-\delta)-\sqrt{(\alpha-\delta)^2+4\beta\gamma}}{2\beta}.$$

We shall take as general case, that β, γ, and δ are all complex; then λ, p, and q will also be complex.

$$f(z) = \frac{1}{\lambda} log \frac{pz+1}{qz+1} = \frac{1}{\lambda} log \frac{p}{q} + \frac{1}{\lambda} log \frac{z+p^{-1}}{z+q^{-1}} = \frac{1}{\lambda} log \frac{p}{q} + \frac{1}{\lambda} log \frac{z_0+p^{-1}}{z_0+q^{-1}} + n.$$

From

$$log \frac{z+p^{-1}}{z+q^{-1}} = log \frac{z_0+p^{-1}}{z_0+q^{-1}} + \lambda n \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

ensues that for an infinite value of n the point z takes either the value $-p^{-1}$ or the value $-q^{-1}$. We shall call the points $z = -p^{-1}$ and $z = -q^{-1}$ the *limiting points* and we shall put $-p^{-1} = g'$, $-q^{-1} = g''$.

Thus our equation (2) becomes

$$log \frac{z-g'}{z-g''} = log \frac{z_0-g'}{z_0-g''} + (\mu + i\nu)n, \quad . \quad . \quad . \quad . \quad (3)$$

where we have replaced λ by $\mu + i\nu$.

Let us choose g' and g'' as auxiliary origins and let us call

$$z - g' = z' = \varrho' e^{i\vartheta'} \quad , \quad z - g'' = z'' = \varrho'' e^{i\vartheta''},$$

we then find out of (3)

$$log \frac{\varrho'}{\varrho''} + i(\vartheta' - \vartheta'') - log \frac{\varrho'_0}{\varrho''_0} - i(\vartheta'_0 - \vartheta''_0) = \mu n + i\nu n,$$

where by separating the real part from the imaginary we find

$$log \frac{\varrho'}{\varrho''} - log \frac{\varrho'_0}{\varrho''_0} = \mu n \quad , \quad (\vartheta' - \vartheta'') - (\vartheta'_0 - \vartheta''_0) = \nu n, \quad . \quad . \quad (4)$$

or

$$\frac{\varrho'}{\varrho''} = \frac{\varrho'_0}{\varrho''_0} e^{\mu n} \quad , \quad \theta' - \theta'' = \theta'_0 - \theta''_0 + \nu n. \quad . \quad . \quad . \quad . \quad (5)$$

Elimination of n leads, when μ and ν are neither of them equal to zero, to

$$\varrho' e^{-\frac{\mu}{\nu}\theta'} : \varrho'' e^{-\frac{\mu}{\nu}\theta''} = \varrho'_0 e^{-\frac{\mu}{\nu}\theta'_0} : \varrho''_0 e^{-\frac{\mu}{\nu}\theta''_0} = C. \quad . \quad . \quad (6)$$

By putting

$$\varrho' = c' e^{\frac{\mu}{\nu}\theta'} \quad , \quad \varrho'' = c'' e^{\frac{\mu}{\nu}\theta''} \quad , \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

we find

$$c' = C c''. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

The equations (7) and (8) determine together a so-called *logarithmic double spiral* [1]), with the points g' and g'' as poles.

From the second equation (5) ensues that the angle $\theta' - \theta'' = \varphi$ between the two auxiliary radii vectores $g'z$ and $g''z$ increases arithmetically uniformly, whilst the first equation (5) shows us that the quotient of the auxiliary radii vectores increases geometrically uniformly.

For the case α, β, γ, and δ real, some simplifications appear.

We shall distinguish three cases.

A. $(\alpha - \delta)^2 + 4\beta\gamma > 0$, $\alpha\delta - \beta\gamma < 0$.

The quantities p and q are real, so the points g' and g'' lie on the real axis. Farthermore we have $e^{\lambda} < 0$, so that $\nu = \pi$.

Hence the orbit of z is a *logarithmic double spiral*, whose two poles lie on the real axis.

A special case is furnished by the condition $\alpha + \delta = 0$, or $\mu = 0$.

From the first equation (5) now ensues that the quotient of the auxiliary radii vectores is constant, so that the point z describes a circle of APOLLONIUS of the triangle $g'g''z_0$, whilst the angle $g'zg''$ increases uniformly with n. An example of the latter case is furnished by $y = \frac{1}{x}$; here $g' = +1$, $g'' = -1$.

B. $(\alpha - \delta)^2 + 4\beta\gamma > 0$, $\alpha\delta - \beta\gamma > 0$.

The points g' and g'' lie on the real axis, whilst $e^{\lambda} > 0$, thus $\nu = 0$.

Now the second equation (5) shows us, that $\theta' - \theta'' = \varphi$ is constant, so that the point z describes the circle passing through g', g'' and z_0.

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[1]) For the logarithmic double spirals the reader may consult: HOLZMÜLLER, Ueber die logarithm. Abbildung etc Zeitschr. f. Math. u. Physik., Vol. 16. (1871), p. 281.

All ∞^2 initial points z_0 furnish thus together all circles of the pencil of which g' and g'' are the base points.

Let us suppose point z determined on its orbit as point of intersection of this orbit with an element of the conjugated pencil of circles, intersecting the real axis a. o. in a point s, then evidently $g'z : g''z = g's : g''s$ holds, so that the equation (5) expresses that the quotient $g's : g''s$ increases geometrically uniformly. (This property enables us to construct easily the points z belonging to given values of n). Farthermore holds $g' = z_{-\infty}$ and $g'' = z_{+\infty}$.

C $(\alpha - \delta)^2 + 4\beta\gamma < 0$.

The points g' and g'' lie symmetrically with respect to the real axis, p and q being conjugate complex. As mod. $e^{\mu i} = 1$ we have $\mu = 0$. The ratio $\varrho' : \varrho''$ is now constant, so that the point describes a circle of APOLLONIUS of $\Delta g' g'' z_0$, i.e. a circle of the pencil with g' and g'' as point circles. We can again regard the point z as if originated by intersection of the orbit with a circle of the conjugated pencil of circles. As the angle $g'zg''$ increases uniformly with n we can easily construct with the aid of the conjugated pencil of circles the points z belonging to definite values of n. It is clear that the orbit of z when n increases indefinitely is described innumerable times, so that the function $\varphi_n(x)$ has as a function of n a real period. If ν is commensurable with π, then this period is a mensurable number.

If particularly $\alpha + \delta = 0$ holds, then $\nu = \pi$. This case is a. o. realized in the function $y = \frac{-1}{x}$; here $g' = i$, $g'' = -i$.

V. $y = \frac{\alpha x + \beta}{\gamma x + \delta}$, where $(\alpha - \delta)^2 + 4\beta\gamma = 0$.

Here we are in the *parabolic* case.

$$f(x) = \frac{\alpha x + \beta}{\frac{\alpha - \delta}{2} x + \beta},$$

or, if we put $-\frac{\beta}{\alpha} = a$, $\frac{-2\beta}{\alpha - \delta} = \frac{\alpha - \delta}{2\gamma} = g$,

$$f(x) = \frac{2\alpha}{\alpha - \delta} \cdot \frac{x - a}{x - g},$$

so

$$f(z) = \frac{2\alpha}{\alpha - \delta} \cdot \frac{z - a}{z - g} = \frac{2\alpha}{\alpha - \delta} \cdot \frac{z_0 - a}{z_0 - g} + n,$$

so that

$$\frac{z-a}{z-g} = \frac{z_0-a}{z_0-g} + \frac{a-\delta}{2\alpha} \cdot n = \frac{z_0-a}{z_0-g} + (\mu+iv)n. \quad . \quad . \quad . \quad (9)$$

The difference between our method and that of KLEIN arises from the fact that KLEIN allows the quantity $\frac{a-\delta}{2\alpha} \cdot n$ to increase *really*.

If we take a and g as auxiliary origins and if we put

$$z-a = z' = \varrho' e^{i\theta'}, \quad z-g = z'' = \varrho'' e^{i\theta''},$$

then the equation (9) takes the form of

$$\frac{\varrho'}{\varrho''} e^{i(\theta'-\theta'')} = \frac{\varrho'_0}{\varrho''_0} e^{i(\theta'_0-\theta''_0)} + (\mu + iv)\, n$$

or

$$\frac{\varrho'}{\varrho''}\{\cos(\theta'-\theta'') + i\sin(\theta'-\theta'')\} = \frac{\varrho'_0}{\varrho''_0}\{\cos(\theta'_0-\theta''_0) + i\sin(\theta'_0-\theta''_0)\} + (\mu+iv)n,$$

from which ensues

$$\left.\begin{aligned} \frac{\varrho'}{\varrho''}\cos(\theta'-\theta'') &= \frac{\varrho'_0}{\varrho''_0}\cos(\theta'_0-\theta''_0) + \mu n, \\ \frac{\varrho'}{\varrho''}\sin(\theta'-\theta'') &= \frac{\varrho'_0}{\varrho''_0}\sin(\theta'_0-\theta''_0) + vn. \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad (10)$$

If we put $\mu = \sigma\cos\tau$, $v = \sigma\sin\tau$ $\left(\text{i. o. w. } \frac{\alpha-\delta}{2\alpha} = \sigma e^{i\tau}\right)$ we find out of (10) when eliminating n:

$$\frac{\varrho'}{\varrho''}\sin(\theta'-\theta''-\tau) = \frac{\varrho'_0}{\varrho''_0}\sin(\theta'_0-\theta''_0-\tau) = c. \quad . \quad . \quad . \quad (11)$$

It is clear, that the orbit as found by KLEIN follows from ours by putting $\tau = 0$. The orbit of KLEIN can thus serve as iteration-orbit for real values of the quantity $\frac{\alpha-\delta}{2\alpha}$, thus of $\frac{\delta}{\alpha}$.

To investigate the curve determined by the equation (11) we imagine the circle passing through g and a and of which the arc ga amounts to 2τ, so that from each point of the supplementary arc the line ga is seen under the angle τ. (See fig. p. 511).

If we connect g with z_0 and z, the connecting lines will meet the circle in m_0 and m.

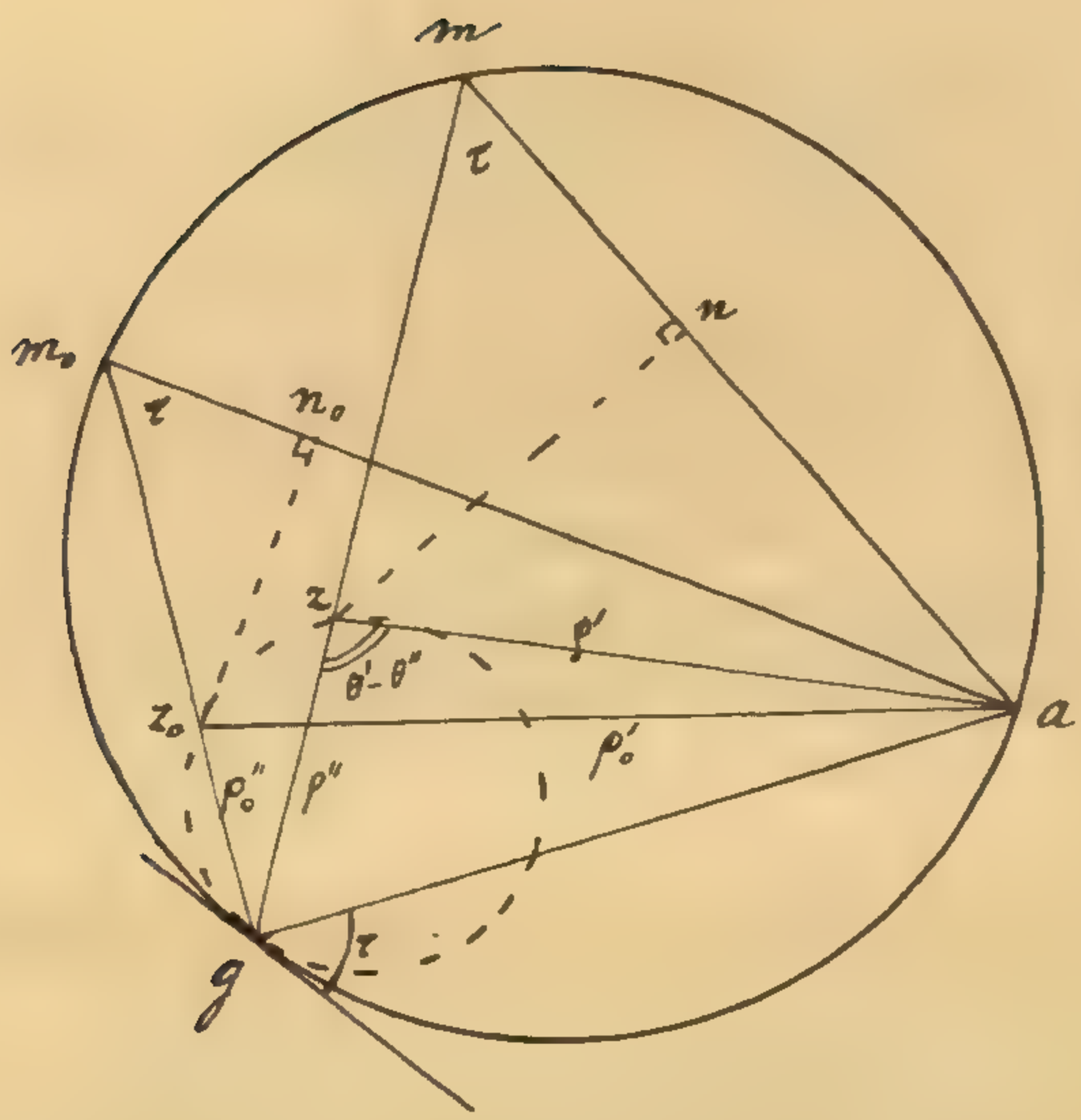
Now $\angle gma = \angle gm_0a = \tau$

Farthermore $\angle zam = \theta' - \theta'' - \tau$, $\angle z_0am_0 = \theta'_0 - \theta''_0 - \tau$.

If we let fall the normals z_0n_0 and zn on am_0 and am, then $z_0n_0 = \varrho'_0\sin(\theta'_0 - \theta''_0 - \tau)$ and $zn = \varrho'\sin(\theta' - \theta'' - \tau)$.

The equation (11) now demands

$$\frac{zn}{zg} = \frac{z_0n_0}{z_0g} \quad \text{or} \quad \frac{zg}{z_0g} = \frac{zn}{z_0n_0} = \frac{zm}{z_0m_0}.$$



It is therefore evident that we arrive from points m to points z by diminishing or enlarging the chords gm in a definite ratio.

So the orbit of z is a circle touching the auxiliary circle (m) in g, whose tangent in g forms in that way the angle τ with the line ga.

If the quantities α, β, γ, and δ are real, then a and g are real, whilst $\nu = 0$, therefore also $\tau = 0$. The points a and g therefore lie on the real axis and the orbit of z touches the real axis in the point g. If on the other hand $\mu = 0$, then the centre of the orbit lies on the line ga.

The way in which z changes with n we can read from the equations (10).

If we suppose the point z to be furnished by the circle, which passes through g and z and whose centre lies on ga, then the first equation (10) tells us that the *reciprocal* value of the radius of that circle increases arithmetically uniformly that i. o. w. the radii of the circles through g whose centres lie on ga and which pass through z_1, z_2 etc., form an *harmonic series*. If on the other hand we suppose that the point z is constructed as point of intersection of its orbit with the circle through z touching the line ga in g, it then follows easily out of the second equation that also the *reciprocal* value of the radius of this circle increases arithmetically uniformly, that i.o.w. the radii of the circles touching ga in g and passing through the points z_1, z_2 etc. form an *harmonic series*.

It is clear that for the case α, β, γ, and δ real, thus $\nu = 0$ and $\tau = 0$, only the first determination of the course of z can serve,

whilst in the case $\mu = 0$ only the second determination retains its validity.

VI. $y = x^{\alpha}$, $y_n = x^{\alpha^n}$, $f(x) = \dfrac{log\ log\ x}{log\ \alpha}$.

$$f(z) = \frac{log\ log\ z}{log\ \alpha} = \frac{log\ log\ z_0}{log\ \alpha} + n.$$

Let us put $log\ \alpha = \mu + i\nu$, we then have

$$log\ log\ z = log\ log\ z_0 + (\mu + i\nu)\, n,$$

so

$$log\ z = log\ z_0 \,.\, e^{\mu n}\,(cos\ \nu n + i\ sin\ \nu n),$$

or

$$log\ \varrho + i\theta = (log\ \varrho_0 + i\theta_0)\, e^{\mu n}\,(cos\ \nu n + i\ sin\ \nu n), \quad . \quad . \quad (12)$$

from which ensues

$$\left.\begin{aligned} (log\ \varrho)^2 + \theta^2 &= \{(log\ \varrho_0)^2 + \theta_0^{\,2}\}\, e^{2\mu n}, \\ \frac{log\ \varrho}{\theta} &= \frac{log\ \varrho_0\ cos\ \nu n - \theta_0\ sin\ \nu n}{log\ \varrho_0\ sin\ \nu n + \theta_0\ cos\ \nu n}. \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad (13)$$

Out of these equations follows by elimination of n the orbit of z. For the case α positive, so $\nu = 0$, the second equation passes into

$$\frac{log\ \varrho}{\theta} = \frac{log\ \varrho_0}{\theta_0} = \varkappa,$$

or

$$\varrho = e^{\varkappa\theta}.$$

The orbit of z is in this case a logarithmic spiral around the origin, which is *independent* of α.

If $mod\ \alpha = 1$, then $\mu = 0$, so that the first equation (13) tells us that

$$(log\ \varrho)^2 + \theta^2 = (log\ \varrho_0)^2 + \theta_0^{\,2} = \tau^2$$

or

$$\varrho = e^{\pm\sqrt{\tau^2 - \theta^2}}.$$

This curve is likewise independent of the argument of α.

The function $y = x^{-1}$, which we have regarded on one hand under IV *A*, $\mu = 0$, and which then furnished for the orbit of z a circle, we can also range under the case treated last. If namely we take $y = x^{-1}$ as a special case of $y = x^{\alpha}$ ($mod.\ \alpha = 1$, $arg.\ \alpha = \pi$), we then find for the orbit of z quite a different curve.

To this remarkable property of $y = x^{-1}$ we hope to refer more explicitly later on.

Physics. — *"On the motion of the bridge of the violin"*. By J. W. GILTAY and Prof. M. DE HAAS. (Communicated by Prof. H. KAMERLINGH ONNES).

(Communicated in the meeting of November 27, 1909).

1. In the following lines an account is given of an experimental research the object of which was to make a contribution to our knowledge of the manner in which the vibrations of the strings are transmitted to the roof of a violin by the bridge.

As far as we know the literature on the physics of bow instruments is very limited and leaves the true nature of the motion of the bridge undecided.

HELMHOLTZ [1]) says: "Der eine Fuss des Steges ruht auf einer relativ festen Unterlage, nämlich auf dem sogenannten Stimmstocke, einem festen Stäbchen, welches zwischen der oberen und unteren Platte des Körpers eingebaut ist. Der andere Fuss des Steges allein ist es, welcher die elastischen Holzplatten und mittels deren Hilfe die innere Luftmasse des Körpers erschüttert."

From this description cannot be inferred whether the bridge vibrates principally in its own plane i. e. at right angles to the longitudinal direction of the strings, or at right angles to its own plane i. e. in the direction of the strings.

VAN SCHAIK [2]) remarks: "By the vibrations of the bowed string a motion of the bridge is set up which consists in an oscillation about a line parallel to the length of the violin: in this manner the movable foot of the bridge communicates vibration to the roof of the violin and thus to the air." His opinion therefore is that the bridge vibrates in its own plane perpendicularly to the direction of the strings.

APIAN-BENNEWITZ [3]) observes: "dass nämlich der rechte Fuss eine viel geringere Bebung als der linke zu machen hat und dass die Thätigkeit des linken Fusses als eine hämmernde zu bezeichnen ist." His view is thus the same as VAN SCHAIK's, as appears further from page 133 of his book.

BARTON [4]) in conjunction with GARRET and afterwards with PENTZER has

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[1]) Tonempfindungen, 3e Ausg. p. 146.

[2]) Dr. J. BOSSCHA, Leerboek der Natuurkunde, III, bewerkt door Dr. W. C. L. VAN SCHAIK, 5th Ed., p. 170.

[3]) Die Geige, der Geigenbau und die Bogenverfertigung. WEIMAR, BERNHARDT FRIEDRICH VOIGT, 1892, p. 125.

[4]) Philosophical Magazine, 6th Series, Vol X, XII and XIII.

investigated the nature of the vibrations of the string, bridge, and roof of a sonometer as also of the air inside the sonometer. He examines both motions of the bridge and finds that for the same point of the bridge the displacement by the horizontal motion, i. e. in the direction of the string, is about 17 times the amplitude of the vertical motion [1]). As the bridge of the sonometer is entirely different in shape from the bridge of the violin and the sonometer is moreover not fitted with a sound bar, the results of the investigation are not immediately applicable to the motion of the bridge of the violin.

SAVART [2]) in his very important memoir on string instruments does not refer to the motion of the bridge.

2. It seemed to us a priori somewhat improbable that as VAN SCHAIK and others suppose a comparatively massive object like the bridge by vibrating as a whole in its own plane about one of its corners should be able to follow completely the intricate motions of the strings and communicate them to the roof of the violin. It seemed to us more probable that, as BARTON found for the sonometer, both motions should be taken into account.

In order to investigate this experimentally we proceeded as follows.

Fig. 1 represents a violin-bridge manufactured by the well known makers CARESSA & FRANÇAIS of Paris. Fig. 2 shows a small

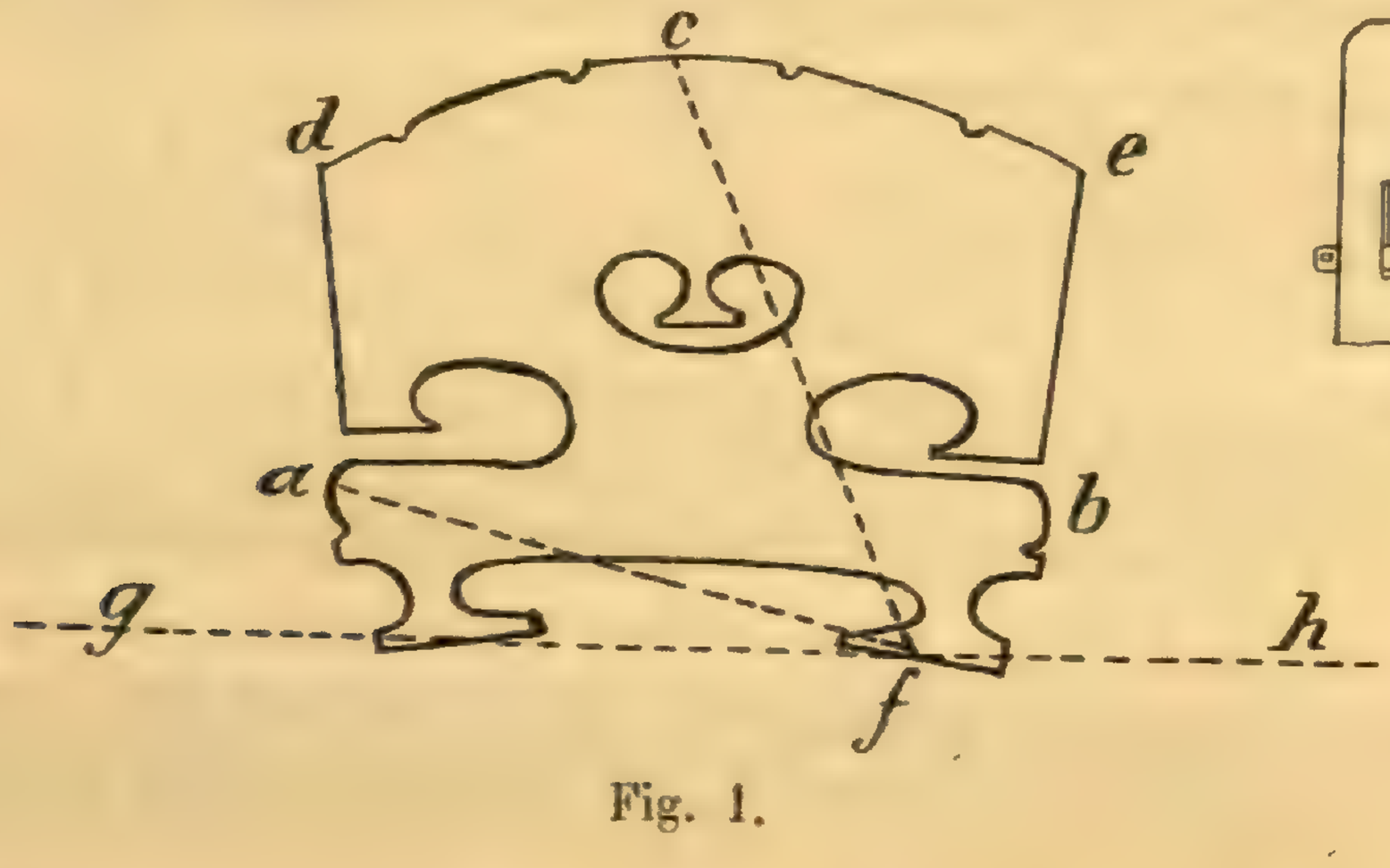

Fig. 1.

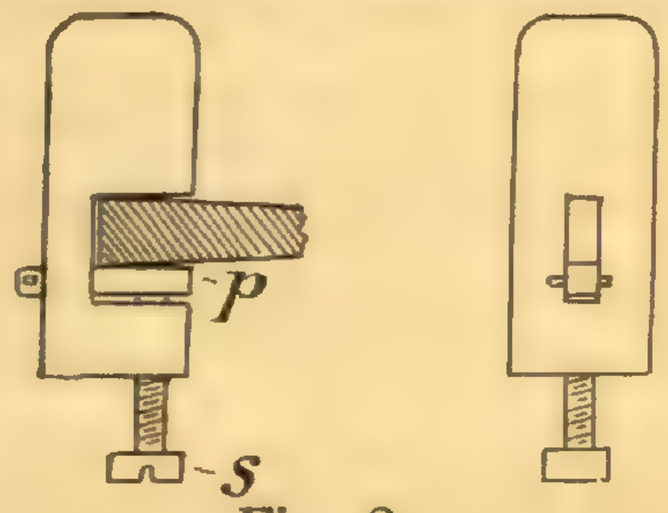

Fig. 2.

metal clamp which can be attached to the bridge at different points. In order not to damage the bridge the screw *s* does not press

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[1]) Phil. Mag. Ser. 6, Vol. XIII, p. 451.

[2]) "Mémoire sur la construction des instruments à cordes et à archet." A reprint of this paper is to be found in: "Nouveau Manuel complet du luthier", by MAUGIN and MAIGNE. Paris, librairie encyclopédique de RORET, 1894, p. 333—398.

directly against the bridge but against a moveable piece of steel *p* The weight of the clamp is rather more than 7 grammes.

If the bridge swings in its own plane about its right foot *f*, then when we attach the clamp to the bridge at *a*, the moment of inertia of the bridge about the axis of rotation at *f* perpendicular to the plane of the bridge will be much increased.

On the other hand when we fix the clamp at *b*, the effect on the moment of inertia will be much smaller.

We found however that there was very little difference in the sound of the violin in the two cases. By fixing the clamp at *a* some damping influence was noticeable in the *g* string; at *b* the *e* string was somewhat damped.

In view of the effect of the clamp being about the same in both cases it is difficult to conclude that the bridge swings principally in its own plane about one of its feet. Moreover the influence of the damper was in *both* cases very small.

The following experiment speaks even more clearly.

The distance between the middle of the right foot and the middle of the upper edge of the bridge *fc* is in our case 38 mms. The distance *fa* is 37 mms.

When the clamp is placed at *c* a strongly damped sound is obtained: this is the well known mute-effect, but even stronger in our case than with the ordinary mute which weighs only about 4 grammes as against ours which weighs over 7 grammes. At *a* the effect is as we saw, extremely small.

As *fc* and *fa* are approximately equal, the increase of the moment of inertia of the bridge is about equal in both cases. If the sound were transmitted by the bridge chiefly by its vibrations about an axis at *f*, the damping effect of our clamp should be about equal in both positions.

As this appears not to be the case we cannot but infer from these experiments that the motion of the bridge in its own plane is not of primary importance for the transmission of the vibrations of the strings to the roof of the violin.

We subjoin as an instance some results obtained by two independent observers each playing his own violin.

Violin with strong sound, about 50 years old, maker unknown, model MAGGINI, very large.

Old violin by a pupil of STAINER's, small strongly arched model, fine mellow sound, but not strong in tone, *d* string least fine, *a* string by far the best, *e* also very good.

Metal damper at *a*. (Fig. 1).

Some damping effect, especially on the *g* string. Rather strong nasal sound.

g string much less fine than without damper.
d harder and inferior.
a inferior.
e improved.
none of the strings damped, respond as promptly as without.

Metal damper at *b*.

Some damping effect, especially on the *e* string. *e* better than usual.

g string better than usual.
d „ worse „ „
a „ „ „ „
e „ „ „ „
g, *d* and *a* respond more promptly than otherwise. The *e* string is slightly damped.

Metal damper at *c*.

Damping much stronger than at *a*. Effect the same as with a mute, only less good than with an ordinary mute.

Mute effect on all strings, but much more strongly damped than with the ordinary mute.

It will be seen that the two observers agree entirely as regards the main effect: the damper at *c* gives the ordinary mute effect. At *a* and *b* the effect is absent or at least only very small; again both observers find the effect of placing the clamp at *a* about the same as at *b*.

The small differences in the results of the two observers may be due to individual differences but also to the great difference between the two instruments.

The following observations prove also, that the parallel motion of the bridge has little influence in the transmission of the string motion to the roof of the violin.

The observers and violins were the same as in the previous experiments and the same damper of 7 grammes was used.

Metal damper at *d*. (Fig. 1.)

Mute effect, strongest on the *g* side.

g string strongly damped.
d string less, bad in tone.
a string still less, bad.
e „ „ „ „

Metal damper at *e*.

Damping, diminishing towards the *g* side. The *g* string has retained its original tone better than the *e* string in the *d* position of the damper.

e damped, but much less than the *g* in the *d* position of the damper.
a less damped.
d damped, gives the mute-sound more than the *a* string, but is still comparatively strong in tone.
g less damped than *d*, very ugly.

Both observers thus found, that in the position *d* the damping effect diminished towards *e* and vice versa.

Thus e. g. in the *e* position of the damper the *g* string was but little damped, although in this case assuming the bridge to vibrate chiefly in its own plane, the *g* string would act on a bridge with much increased moment of inertia which would involve strong damping.

Fig. 3.

We think therefore that we may infer from these experiments that the motion of the bridge does not principally take place in its own plane about one of its feet, but that it vibrates chiefly transversely, as shown diagrammatically in Fig. 3 where *ab* represents the bridge in section. On this assumption the results of all the above experiments are completely explained:

I. A damper placed at *a* has much less damping influence than a damper at *c*, as the moment of inertia about *gh* is much less increased in the former case.

II. The effect is about the same whether the damper is attached at *a* or at *b*. It is clear that the moment of inertia of the bridge, with the clamp attached, about *gh* has about the same value in the two cases.

III. Again the results of the second set of experiments become

intelligible when a transverse vibration of the bridge is admitted: we found in that case that the damping effect diminished towards the right when the clamp is fixed at d and vice versa. By weighting the bridge at the top corners the vibration is no longer symmetrical; the part which is loaded at the top will vibrate less strongly than the unloaded part.

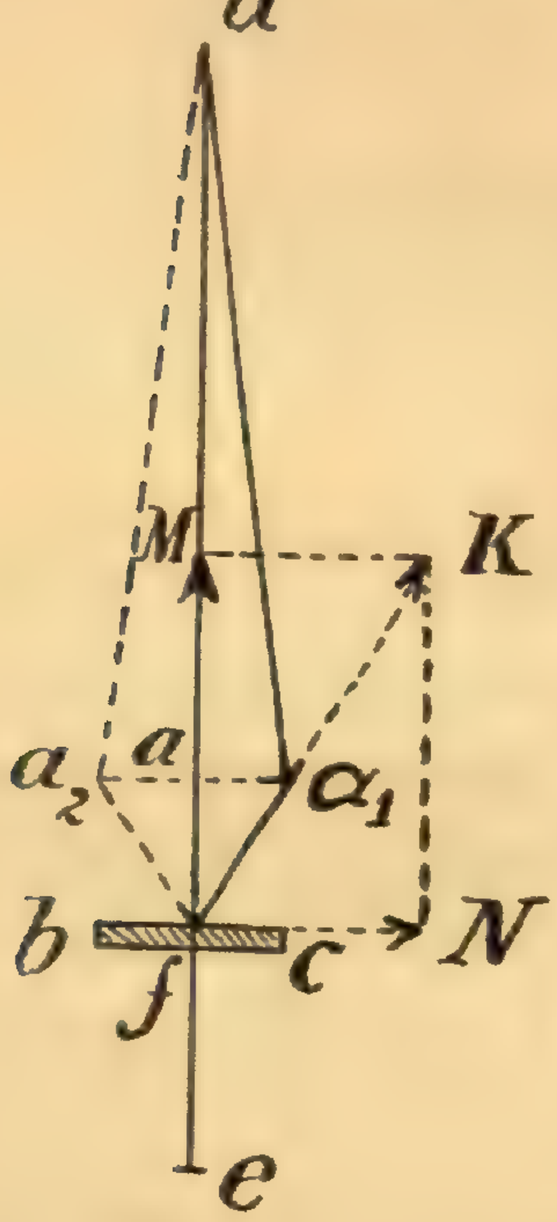

Fig. 4.

3. An additional question with regard to the two motions of the bridge suggested itself in the investigation. In fig. 4 de represents the string at rest, bc the bridge: when the string is deflected to the right (da_1f), the tension K of the string has a component M at right angles to the plane of the bridge and a component N in the plane of the bridge. When the string has its greatest deviation to the left, the component M has the same direction as before, the component N the opposite. It follows that the bridge completes two vibrations in the direction of the string to one vibration of the string itself, whereas the motion parallel to the bridge has the same period as the string.

The sound of the violin is produced almost exclusively by the vibration of the roof; the string by itself imparts but a very small amount of energy to the air directly. If we suppose that the sound given by the string directly may be neglected in comparison to the much stronger sound which is due to the roof, and that the effect of the parallel motion of the bridge may also be neglected as against the much greater effect of the transverse motion, all the notes of the violin should be an octave higher than the pitch of the string, assuming that the strings deviate on both sides of the position of equilibrium.

The correctness of this conclusion however did not seem to us very probable: presumably if real, this striking fact would have been observed and communicated by previous observers.

We have therefore investigated the question experimentally by putting a steel string on a violin and making it vibrate electromagnetically.

We took a steel guitar string and put it in the position of the d string. Close to it a small electromagnet of the ROMERSHAUSEN type was fixed in a stand about vertically above the string, near the place where it is usually bowed. The coil of the electromagnet was in

circuit with three accumulators and a KÖNIG electromagnetic tuning fork ($Fa_3 = 682$ v. s.). The fork was placed in a distant room. The tension of the string was regulated until the violin when the string was bowed gave a note slightly lower than the fork. The fork was then started and the note of the string raised by pressing it with the finger until no beats were heard.

The note given out by the violin was now unmistakably Fa_3.

Now if there really were a difference of an octave between the note of the violin (Fa_3) and the note of the string itself, the string ought under the influence of the electromagnet to have given the note Fa_2. This is however impossible: an electromagnet magnetised by a fork Fa_3 can produce in a string the notes Fa_3 Fa_4, Fa_5 etc. but never the note Fa_2. The experiment was thus by itself sufficient to show that the note given by the violin has the same pitch as the note of the string itself, even when the excursions of the string on the two sides of its position of equilibrium are about equal.

Thinking that the octave might perhaps appear, if the parallel motion of the bridge were damped down, we loaded the left foot of the bridge with our metal clamp, but even then the octave could not be heard.

As the question seemed to us of great importance we tried to solve it in a different more direct manner by an experiment in which the sound of the string was heard by itself.

On a heavy zinc-block of 80 by 40 cms and 3 $^1/_2$ cms thick (Fig. 5), two metal bridges are fitted (Fig. 6) at a distance from each other of 32$^1/_2$ cms. An a-string 0,7 to 0,75 mm thick was tied

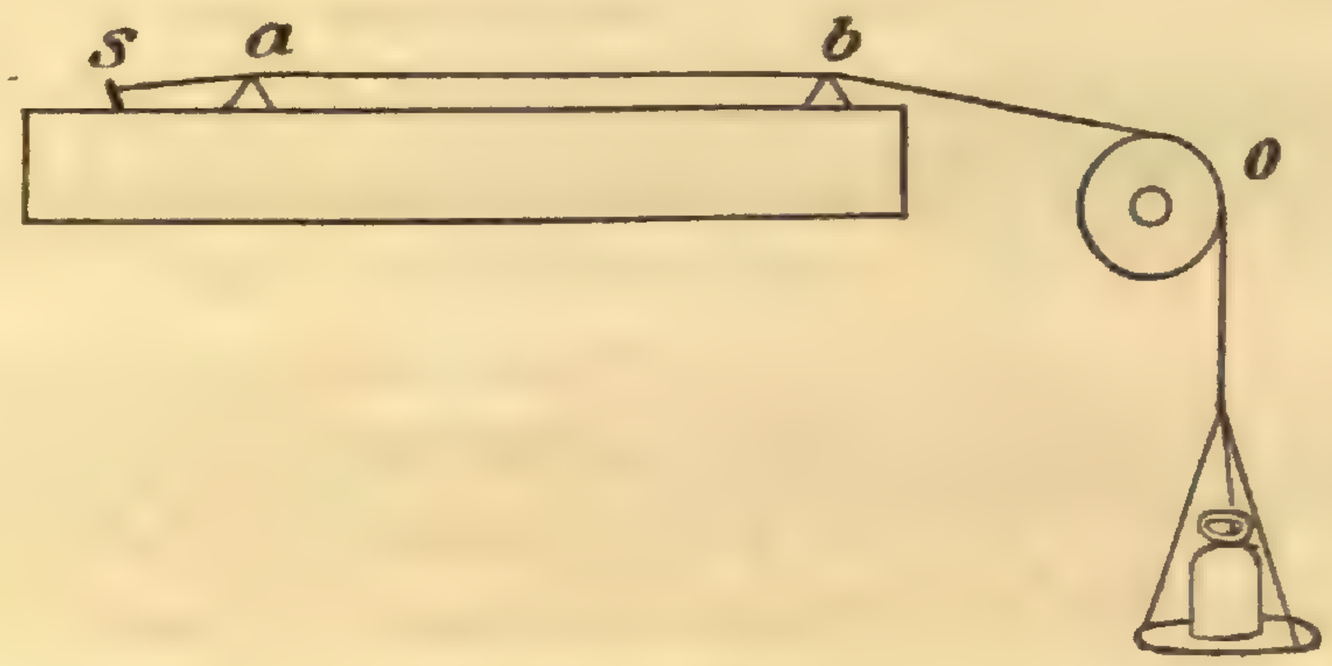

Fig. 5.

to a pin s, the other end being attached to a cord going over a pulley and a pan weighted with 6 kilogrammes. When bowed the string sounded a note near Ut_4. The friction of the string on the bridges and of the cord on the pulley enabled us to slightly alter

Fig. 6.

the pitch by turning the wheel of the pulley. In this manner the string was accurately tuned to Ut_4 (1023,9 v.s), so that it produced no beats in a resonator Ut_4.

Next an a string of the same thickness was put on a violin (fig. 7). The distance of a to b was again 32,5 cms. The violin was clamped

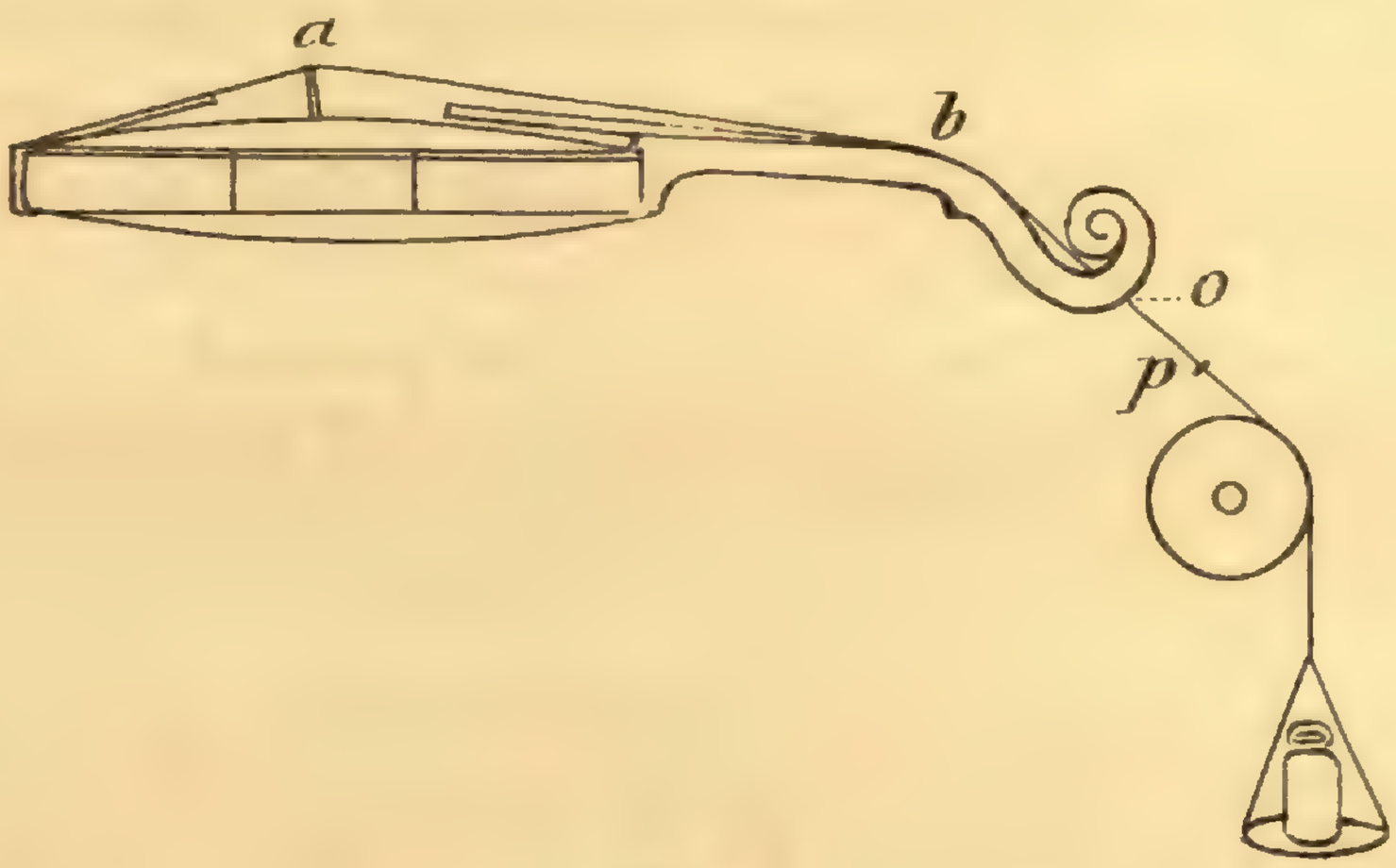

Fig. 7.

on the table with some wooden blocks; in the neck of the violin a hole O was bored, through which the string was made to pass. As the friction of the string on the usual ebony peg would have been too great, a metal peg was substituted which is represented in fig. 8.

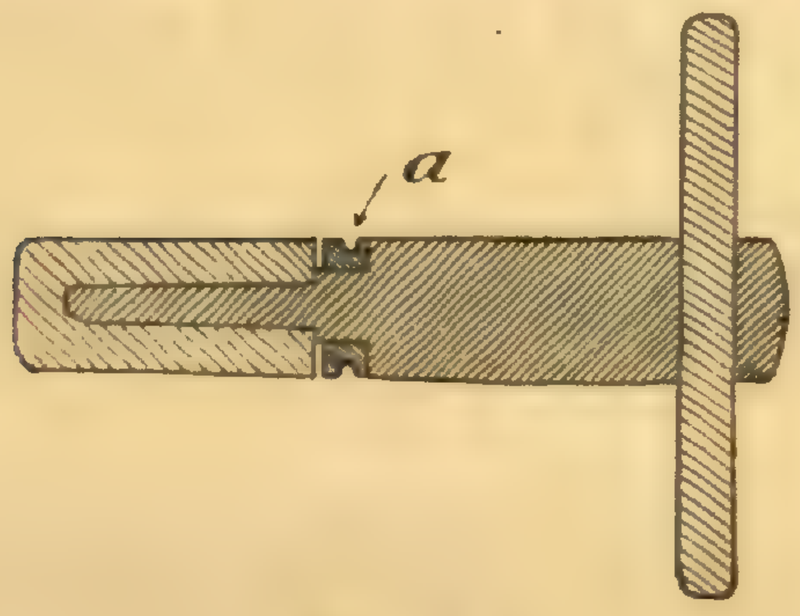

Fig. 8.

The string passes over the small metal wheel a. At p a cord was tied to the string which ran over a pulley and had a pan attached to it. The a string was now stretched by placing weights in the pan until the violin on bowing sounded Ut_4 accurately. It was found that a weight of 6 kilograms was required to do this, i. e. the same as with the zinc block. That the violin and the string on the zinc block gave the same note, i.e. without a difference of an octave, was confirmed not merely by the ear but also by the aid of resonators: the resonator Ut_4 responded to both notes, the resonator Ut_3 did not. If the note given by the violin had been an octave higher than that of the string on the zinc plate (i. e. Ut_5) the resonator Ut_4 would not have responded to the violin note.

We have also determined the note which the string gave at the above tension by calculation.

For that purpose the string was cut at a and b (fig. 7) whereby its length shrunk to 30 cms. The weight of this piece was found to be 0,15 grams.

By substituting in the formula $t = \sqrt{\frac{pl}{gs}}$ where t is half the period, $p = 0{,}15$ gr, $l = 32{,}5$ cms, $g = 981{,}2$ cms sec^{-2}, $s = 6000$ gr, it follows $t = \frac{1}{1099}$ sec.

According to this calculation the string would have a frequency of $\frac{1099}{2} = 549{,}5$ complete vibrations whereas in reality the frequency was 511,9 (Ut_4).

These numbers agree sufficiently to show with certainty that in both cases the fundamental note of the string was heard. The comparatively small difference can be explained by assuming that the tension of the string was not exactly 6000 grams in consequence of the friction of the string on the bridges and of the cord on the pulleys.

From these experiments it appears that in the mixed sound which the violin produces the fundamental note produced by the parallel motion of the bridge and by the motion imparted to the air directly by the string is still present in sufficient intensity to give the sound the character of the fundamental as far as the pitch is concerned. [1])

It is indeed well known that the fundamental which determines the pitch of a composite note may be of smaller intensity than the overtones of the mixture, as HELMHOLTZ showed to be the case with the piano. [2])

We thus know that the sound given by a violin must be ascribed to three distinct causes:

a. a vibration imparted to the air by the string.

b. a vibration which the roof of the violin acquires from the parallel swing of the bridge.

c. a vibration communicated to the roof by the transverse vibration of the bridge.

The vibration mentioned under *a* will be left out of account as being of little importance.

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1) Compare RAYLEIGH, "Theory of Sound". second ed. Vol. I p. 208 and BARTON and PENZER, Phil. Mag (6) XIII p. 452.

2) Tonempfindungen, p. 134—135.

If a string is bowed the fundamental of which has a period T, the note will be accompanied by harmonics of periods $^1/_2 T$, $^1/_3 T$, $^1/_4 T$ etc. respectively.

The parallel motion of the bridge will cause a periodical change of pressure of its left foot on the roof of the violin. When the bridge moves to the left the pressure increases and vice versa. The change of pressure may be represented by the following series:

$$a_1 \sin 2\pi \frac{t}{T} + a_2 \sin 2\pi \frac{t}{^1/_2 T} + a_3 \sin 2\pi \frac{t}{^1/_3 T} + a_4 \sin 2\pi \frac{t}{^1/_4 T} \ldots$$

The transverse motion of the bridge will also cause a change in the pressure between the left foot and the roof. When the bridge is pulled forward the front of the left foot will exert a greater pressure on the roof; when the bridge moves back the pressure diminishes. This change of pressure may be represented by a series of the form

$$b_2 \sin 2\pi \frac{t}{^1/_2 T} + b_4 \sin 2\pi \frac{t}{^1/_4 T} + b_6 \sin 2\pi \frac{t}{^1/_6 T} + b_8 \sin 2\pi \frac{t}{^1/_8 T} \ldots$$

As the foot of the bridge has only a small area compared to the large surface of the violin which is set in motion, we may assume that the pressure changes which are due to the parallel and the transverse motions of the bridge respectively, occur *at the same point* of the roof. In order to find the total change of pressure produced by both motions together we must therefore add the two above series. If we assume that the excursion of the roof at the point where the left foot is attached to it is proportional to the change of pressure, the sum of the two series multiplied by a constant will give us the type of motion of the roof at that point.

It is well known that in general a sound becomes mellower according as the partial overtones become weaker and that the intensification of the even overtones especially renders the sound sharper. Many instances of this are to be found in HELMHOLTZ's work already repeatedly quoted (p. 129—133 and p. 151—152). As an illustration of the influence of the overtones on a mixed sound we may also mention the sound of a piano when octaves are played. When an octave is struck on the piano the two notes cannot easily be heard separate, as they can be e. g. with thirds. But only very slight musical training is required to hear in a musical recital that running octaves are played: the sound is then sharper and rougher. The same holds for running octaves on the violin.

When in the above series we diminish the coefficients a_1, a_2, a_3 etc. while leaving the b_2, b_4, b_6 unchanged as far as possible, the

fundamental and odd harmonics are weakened more than the even harmonics. In accordance with the above results of HELMHOLTZ the sound will thereby be made sharper. We have proved this in the following manner by experiment:

To the bridge of a violin at the lowest possible point a metal clamp, represented half size in Figs. 9*a* and 9*b*, was attached. On the left side (i. e. on the side of the *g* string) a copper rod 3 mms

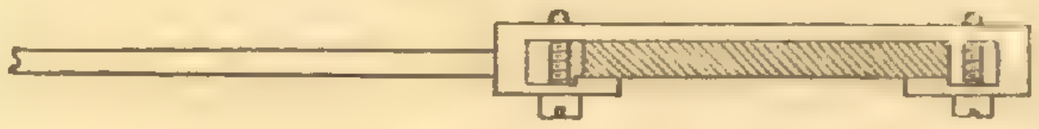

Fig. 9*a*.

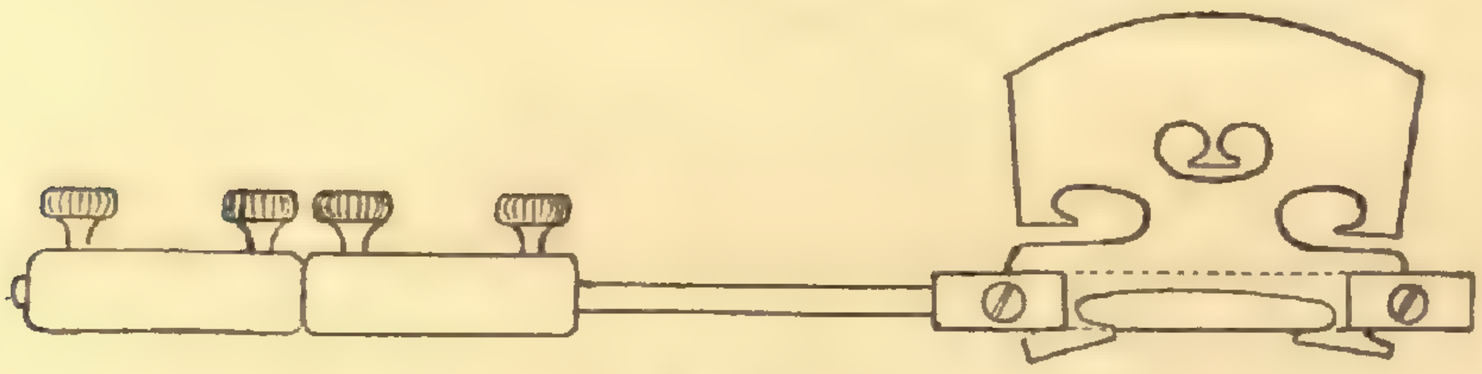

Fig. 9*b*.

thick and 10 cms long was screwed into this clamp. At the end of this rod two ordinary binding screws were fixed, weighing about 18 grammes each.

The moment of inertia of the bridge about the axis through the right foot, perpendicular to the bridge was naturally very much enlarged by these weights. The violin now gave a characteristic nasal sound, especially in the *g* and *d* strings; the timbre resembling most the note of a hautboy. Still notwithstanding the great weakening of the fundamental it continued to impart to the sound the character by which the pitch of a note is distinguished, in other words no change of an octave was perceptible.

When in addition to the clamp shown in Fig. 9*b* the bridge was loaded with two mutes fixed on top of each other and placed on the upper edge of the bridge, the original sound was approximately recovered, as now the transverse as well as the parallel motion of the bridge was damped. Of course the response of the violin at this load was difficult. The two mutes were an ordinary ebony mute with a metal mute, as often used, placed on top.

When a_1, a_2 etc. and b_2, b_4 etc. are all diminished in the same proportion the form of the curve of motion will not change, only the amplitude diminishes: the intensity is weakened, but the timbre remains the same.

If we could diminish the *b*'s and leave the *a*'s unchanged, the sound would become mellower, as in that case only the even upper partials would become weaker, including the first overtone which has the greatest intensity of all.

A mute placed on the bridge damps both motions. But from the fact that it renders the sound mellower we think we may infer that the *b*'s are reduced by it by a higher fraction than the *a*'s.

This would mean that the transverse motion of the bridge is damped to a higher degree by putting on the mute than the parallel motion.

5. We have also tried to show experimentally that the bridge in its parallel motion turns principally about its right foot.

For this purpose we screwed two metal rings into the clamp of fig. 9, which were placed in a horizontal position. The violin was fitted with a steel string, as before moved electromagnetically. While the string was moving a small leaden ball was placed alternately in the two rings; the two balls weighed 34 grms each. They were attached to a thin cord; as nearly as possible at the same moment that one ball was lifted out, the second ball was carefully placed in the other ring. We expected that the sound of the violin would be perceptibly weakened as the ball on the right was removed and the left ball simultaneously put in. But we did not succeed in arriving at a trustworthy result in this manner; in the first place a rattling noise was sometimes apparent while the balls were being exchanged and in the second place the tone of the steel string was not always of the same intensity.

6. The conclusion therefore to be derived from our experiments is that the bridge of a violin performs a parallel as well as a transverse motion and that the timbre of the tone, given by the violin, is modified greatly when the intensity of one of the motions is altered while leaving the other motion unchanged as nearly as possible.

Herewith we have at the same time given the physical explanation of the action of the mute and also of the influence which the use of too thick or too thin a bridge has on the sound of a violin.

The action of the mute is commonly described by calling it "damping" or "deadening" [1]). But if the mute caused nothing but a general damping or reducing of the bridge motion, the mute would only weaken the sound, and the same effect would be obtained by bowing softly on a violin without as by bowing hard on a violin with a mute. That however is by no means the case as every one knows.

Delft, November 1909.

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[1]) Barton. "Textbook on sound", p. 419: "The mute is a small apparatus of wood or metal which fits on the bridge, and thus deadens the sound considerably"

Anatomy. — *"On the slope of the Foramen magnum in Primates"*.

By Prof L. Bolk.

(2nd Paper on the Comparative Craniology of Primates).

In the first paper on the anatomy of the Primate-skull, the position and shifting of the occipital Foramen in Primates was treated. This paper will be devoted more especially to the consideration of the inclination of this plane.

All the writers who have dealt with this subject have pointed out that these two features: position and inclination, stand in a certain relationship to each other, in so far as the closer the Foramen lies to the occipital pole the more vertical a position does it assume, while as it gradually approaches the middle of the cranial base the tendency is towards a horizontal position. This variation in the slope, like the shifting, has been connected with the erect gait of the human body. In the typical quadruped, where the skull more or less hangs from the spinal column, the Foramen lies at the occipital pole of the skull, and the plane is vertical; in human beings, where the longitudinal axis of the body runs vertically, the occipital Foramen lies in the middle of the cranial base, while the plane is almost horizontal. Thus it is seen that this plane is disposed to take up a position perpendicular to the longitudinal axis of the body. Another point of view, first fully developed by Huxley, concerns the connection which is said to exist between the slope of the plane of the Foramen magnum and the degree of prognathism [1]). The more pronounced the prognathism — i. e. the longer the face-skull — the more perpendicular would the Foramen magnum stand. If now a rough comparison be made of an animal's skull with a human skull, the parallelism between these two features is at once noticeable. Huxley, however, believed he could show it even in the skulls of different races of men. From the superposition of the mediagrams of the highly prognathous skulls of an Australian and a Negro on the skull of a Tartar, it was seen that "the plane of the occipital Foramen forms a somewhat smaller angle with the basiscranial axis in those particular prognathous skulls than in the orthognathous".

Welcker [1]) holds a somewhat similar opinion, though he does not express it as being a connection between prognathism and the slope of the Foramen magnum, but between prognathism and the position

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[1]) T. Huxley, On some fossil remains of man. Collected Essays. VII, p. 198.

[2]) H. Welcker, Untersuchungen über Wachsthum und Bau des menschlichen Schädels. Leipzig 1862.

of this opening, which, however, comes practically to the same thing, if a connection between position and slope be assumed. "Biegt am Vorderschädel", he says (l. c. p. 50), "der Oberkiefer des Menschen mehr nach vorn (Prognathismus) so rückt zugleich am Hinterschädel das Foramen medullare mehr nach rückwärts". AEBY[1]) does not agree with HUXLEY: "HUXLEY glaubte die Neigung mit dem Prognathismus in Verbindung bringen zu können. Die Steilheit der Stellung sollte in gleichem Masse wie die letztere wachsen. In unseren Tabellen findet sich keine Bestätigung dieser Ansicht" (l. c. p. 17). AEBY himself sees a connection between the degree of development of the occiput and the slope of the Foramen magnum: "Die Abflächung des Hinterhauptes führt eine Erhöhung des Foramen magnum im Gefolge." This opinion does not really differ in principle from WELCKER's, for if the occiput be markedly flattened the Foramen magnum will lie further back, and thus the opinions of WELCKER and AEBY coincide after all with the opinion already expressed by DAUBENTON, that the For. magn. is the more perpendicular in proportion as it is pushed further backwards. The connection which HUXLEY believed he had shown was, however, of another kind, and AEBY is therefore not correct in representing his opinion as being in contradiction to HUXLEY's. For it is not impossible that the slope is proportional on the one hand to the degree of prognathism, and on the other to the position. Is then the relation between position and slope of such a constancy as TOPINARD made it appear originally when he said[2]): "qu'il suffit de mesurer l'un des deux termes par exemple l'inclinaison du trou occipital pour connaître l'autre, c'est à dire la quantité du déplacement du trou"? This seems à priori improbable, since TOPINARD's method of determining each of the two phenomena possesses merely a very relative degree of accuracy. Indeed TOPINARD himself saw this later[3]), and then expressed himself more cautiously: "Toutefois il n'y a pas un parallélisme rigoureux entre les deux phénomènes."

In general the above writers determined the slope of the Foramen magnum by determining the angle which was formed between the base-line adopted by them, and the line which connects basion and opisthion. The base-line in these researches connected the basion with the nasion or typhlon, and therefore ran through the skull-base. This method has been contested by BROCA, and quite justly, for the size of the angle which is formed by these two lines is

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[1]) C. AEBY, Die schädelformen der Menschen und Affen. Leipzig 1867.
[2]) P. TOPINARD. L'Anthropologie. 4me Edition.
[3]) P. TOPINARD. Éléments d'Anthropologie générale.

not dependent merely on the direction of the Foramen magnum because the direction of the base-line, i. e. one of the legs of the angle, depends on several factors, e.g. the angles of the basis cranii, the length of the skull, the length of the clivus, the position of the nasion, etc. To avoid this difficulty BROCA determined the slope of the For. magn. by an angle made by the plane of this opening with a plane which is entirely independent of the cranial base, viz. that which connects the axes of the two orbitae. He constructed his "angle orbito-occipital" [1]). BROCA here was proceeding from the postulate that the orbital-plane, in Primates at least, is the natural horizontal plane of the skull, as, in the case of normal sight, these animals look straight before them and the orbitae in this circumstance therefore will have the same direction. The correctness of this opinion will be discussed in a following paper.

RAUBER [2]) in a recently published treatise, returns to the old, disused method, takes as base-line again the nasion-basion, and even says that: "eine Beziehung der Neigung des Foramen occipitale auf eine andere Linie als auf die Basallinie führt sehr leicht zu Unverständlichkeiten und entbehrt zugleich der morphologischen Bedeutung".

SCHWALBE, also, lately expressed as his opinion regarding the value of AEBY's base-line as follows: "So rationell auch die von AEBY gezogene Grundlinie ist, ist sie doch nicht geeignet über die Ausbildung der verschiedenen Teile des Schädelraumes Auskunft zu geben." [3])

There is a certain contradictoriness in this criticism. A rational base-line of a craniometrical system must be able to serve as basis for at least a primary division of the skull. I have already briefly stated my objections to base-lines which are drawn through the skull-base, and will come back to this subject in a following paper. Such a line may have a certain value as boundary line between the cerebral- and facial-skull, but as basis of a craniometrical system it is absolutely useless.

HUBER [4]) finally has determined the slope of the For. magn. in Hylobates with regard to the so-called German horizontal, a method which when the skulls to be examined cannot be halved medially is preferable to those of other investigators. The probable error will here be less than by the use of the basal line and certainly likewise less than by employing the horizontal auxiliary line made use of by

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[1]) P. BROCA. Sur l'angle orbito-occipital. Revue d'Anthropologie 1897.

[2]) A. RAUBER. Der Schädel von Kegel. Int. Monatsch. f. Anat. und Phys. 1906.

[3]) G. SCHWALBE. Kritik zu KOHLBRUGGE's: Morphologische Abstammung des Menschen. Globus 11 Juni 1908.

[4]) L. HUBER. Vergleichung des Hylobates und Menschenschädels. München 1902.

LISSAUER [1]), running from the protuberantia occipitalis externa to the point where the ala of the vomer is joined to the rostrum sphenoïdalis.

By the method I have adopted in determining the slope of the For. magn. I have proceeded from the base-line which was described in the first paper, and in so-doing have answered the question as to what angle is made by the plane of the occipital foramen with this line. To express this angle in all Primates always as a positive value it is not possible to measure the angle directly. For in the Primates 3 conditions occur: *a.* the opisthion lies higher than the basion, the For. magn. looks backwards, and the angle is therefore an acute one closed at the back; *b.* basion and opisthion lie at equal distances from the base-line, the For. magn. looks downward, is parallel to the base-line, and the angle $= 0$; *c.* the basion lies higher than the opisthion, the For. magn. looks forward and the angle is an acute one closed in front. To prevent confusion between angles of equal size in cases *a* and *c*, a + or — sign could be used. I think, however, the variations in the inclination might be represented more simply in determining the angle made by the plane of the For. magn. with a perpendicular drawn from the basion to the base-line. In case *a* this angle is always acute, in case *b* it is a right angle, and in case *c* it is an obtuse angle.

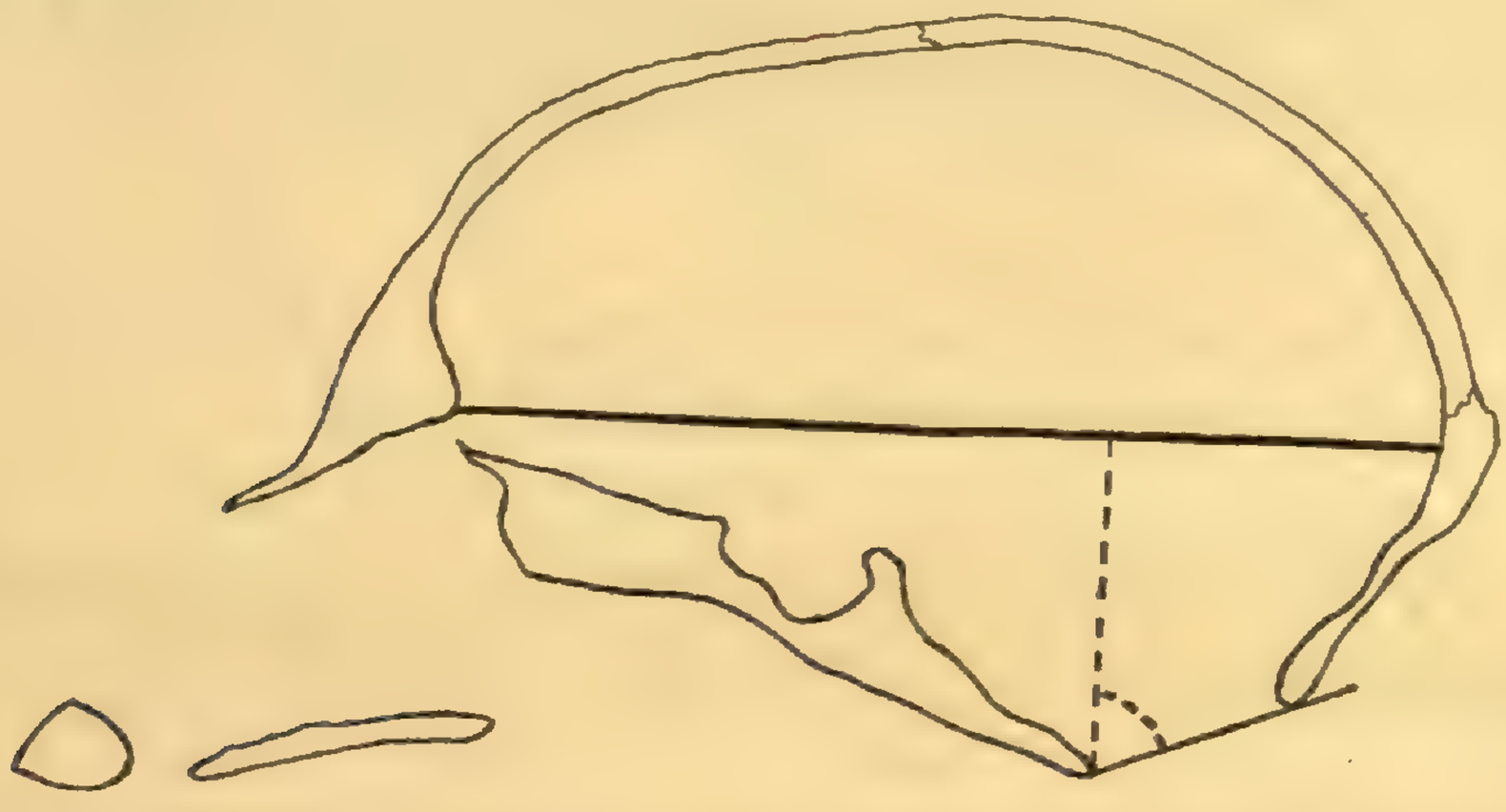

Fig. 1.
Mediagram of an Ateles skull, illustrating the method of determining the slope of the For. magn. ($^3/_4$ natural size).

In Fig. 1 this method is clearly seen on the mediagram of an Ateles skull. The following table gives the results of the researches on the skulls of full-grown monkeys.

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[1]) LISSAUER. Untersuchungen über die sagittale Krümmung des Schädels. Arch. f. Anthrop. XV Bnd. Suppl.

Lemur. 40. Propithecus 42.
Mycetes. (18). 33. 45. 53. 59. Average **47.5**.
Pithecia. 54. 56. 60. 64. Av. **58.5**.
Hapale. 61. 61. 63. 64. 69. 72. Av. **65.**
Chrysothrix. 60. 61. 63. 65. 66. 69. 70. 70. 71. 71. Av. **66.6**.
Cebus. 63. 64. 64. 65. 67. 67. 68. 72. 73. 75. Av. **67.8**.
Ateles. 66. 67. 68. 71. 77. 79. 82. Av. **72.7**.
Cynocephalus. 63. 64. 66. Av. **64.2**.
Inuus. 66. 68. 70. 76. 76. Av. **71.2**.
Macacus ♂. 68. 70. 70. 74. 79. Av. **72.2**.
Macacus ♀. 67. 73. 75. 78. 84. Av. **75.4**.
Cercopithecus. 74. 80. 81. 82. Av. **79.2**.
Colobus. 64. 72. Av. **68**.
Semnopithecus. 60. 61. 61. 64. 68. Av. **62.8**.
Siamanga. 55. 56. 56. 56. 58. 61. 63. 63. 67. 68. Av. **60.2.**
Hylobates. 52. 60. 66. 73. 75. Av. **65.1**.
Chimpanzee. 64. 79. 80. Av. **74.3.**
Gorilla. 63. 63. 66. 70. 76. 77. 80. 80. Av. **71.8**.
Orang. 58. 62. 68. 70. 72. 75. 79. 80. Av. **70.3.**

These figures show in the first place that the slope of the Foramen magnum varies greatly in individual cases, a fact which is apparent by merely looking at the skulls. This individual variability is especially noticeable in the large skulls such as those of the Anthropoids. And yet the general configuration of the skull is but little influenced by these great variations in the slope of the Foramen. As a proof

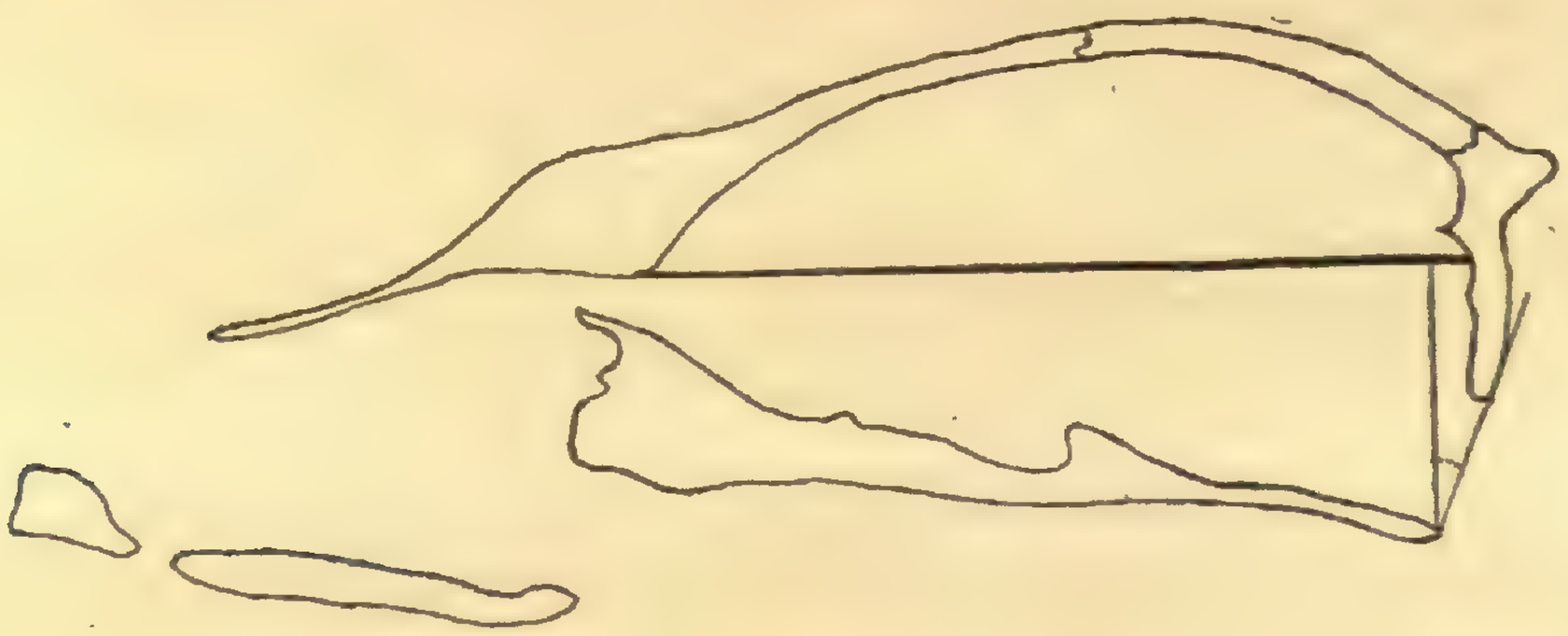

Fig. 2.
Mycetes. ($^3/_4$) Angle of inclination of the For. magn. 18°.

of this, I have given in Figs. 2 and 3 the mediagrams of two Mycetes skulls, with slope-angles of 18° and 59° respectively.

From the figures it can also be seen that a slight shortening of the Clivus is of great influence on the angle of the slope. Now

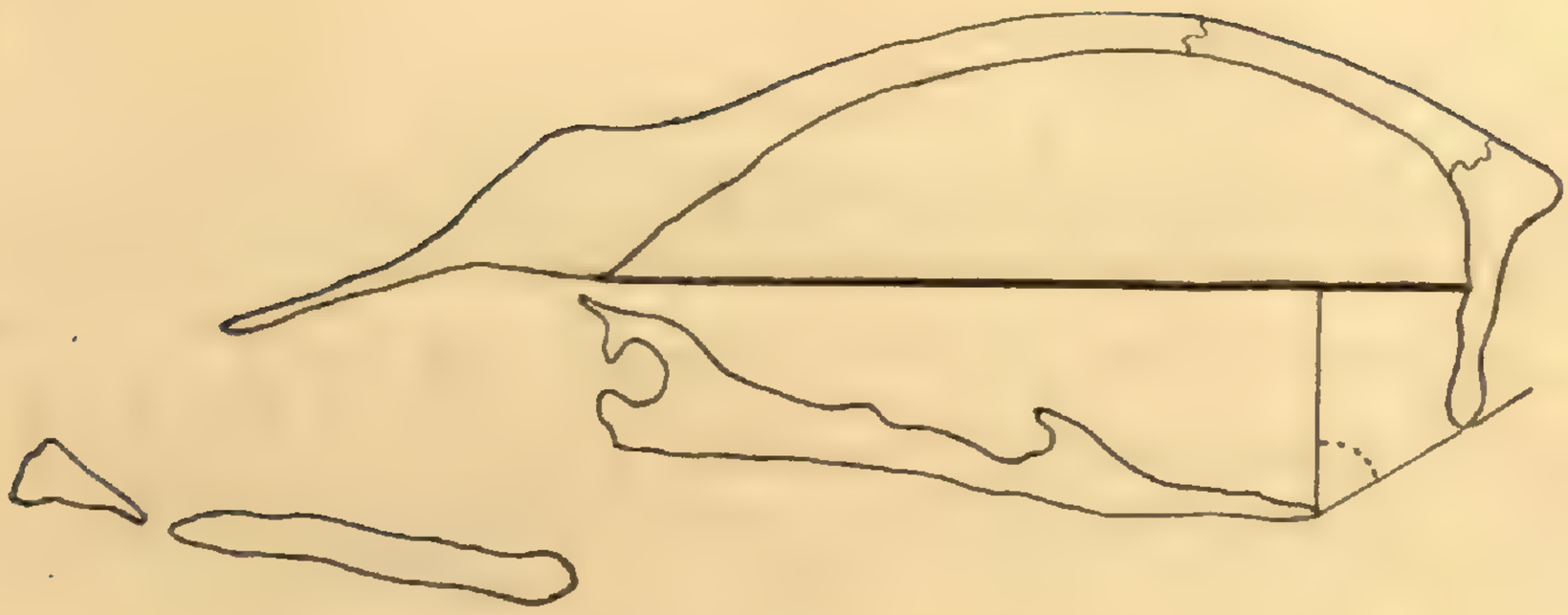

Fig. 3.
Mycetes. ($^3/_4$) Angle of inclination of the For. magn. 59°.

Mycetes occupies a foremost place in the variability of the inclination as in that of the position of the Foramen magnum for reasons fully given in the previous paper. For the other skulls, however, the same holds good. Another cause of the individual variations is the striking difference in sagittal measurement of the For. magnum especially in Anthropoids. In the Orang-outang skulls, for instance, which I used, this measurement varied from 25 to 41 mm.

Nevertheless, in spite of these individual variations some remarkable features are to be detected between the different primate-genera, especially if the series be compared as a whole with one another. It is noticeable that Chrysothrix does not seem to occupy the place attributed to this family in the literature on this subject. Among the Plathyrhines, Cebus, and more especially Ateles, have greater angles, that is to say, in these genera the For. magn. lies more horizontally. In this respect the Chrysothrix is inferior even to most of the families of the Catarrhines. On an external observation, however, the For. magn. seems in this monkey's skull to lie horizontally in consequence of the enormous development of the occiput, and the large share that the squama occipitalis occupies in the formation of the cranial base. (See Fig. 4).

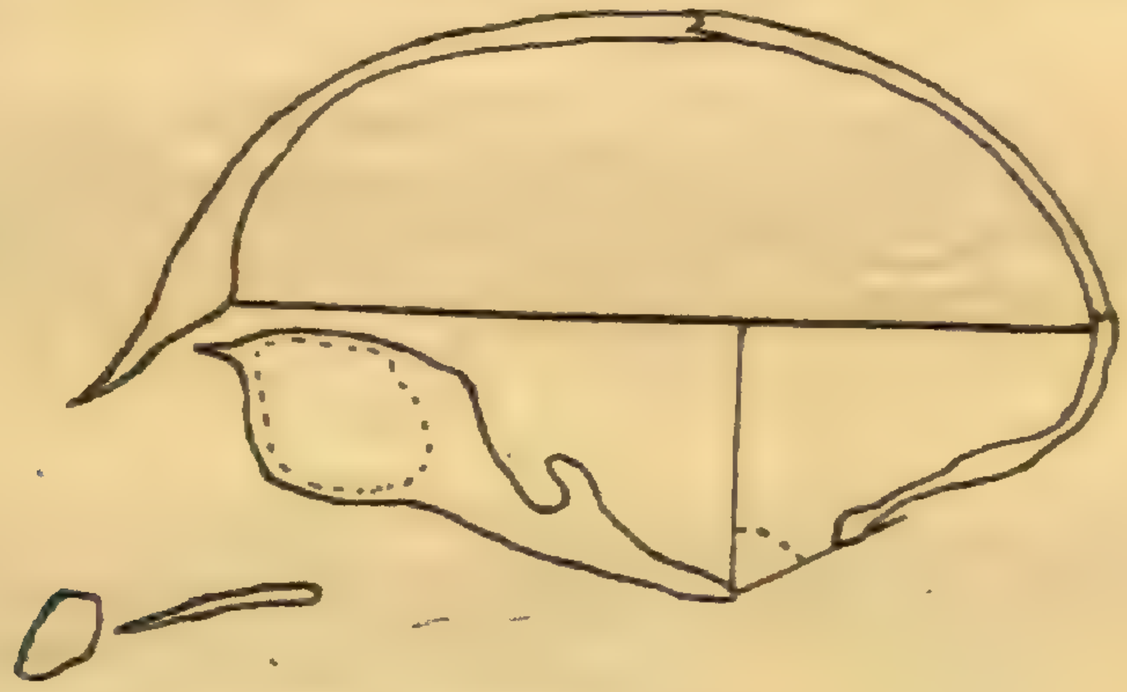

Fig. 4.
Mediagram of the skull of Chrysothrix. ($^1/_1$).

Among the Catarrhine monkeys, the greatest angles, 80° and more, occur among the Anthropoids and the genus Cercopithecus. This genus thus, also as regards the slope of the Foramen magnum, takes the high place which we have already awarded to it in the previous paper on account of the position. And similarly the genus Siamanga takes again the lowest place among this group of Primates. In this otherwise so highly developed monkey the Foramen magnum is inclined more vertically than in any other family of monkeys of the Old World, although it is closely followed by the genus Semnopithecus. A study of the skull base will afford us the opportunity of pointing out more particularly what a quite distinct place the Siamanga takes in the group of Primates, as regards the general form of the cranium. In the first paper I have already mentioned that it is difficult to believe that original conditions have been here persistent.

In the foregoing paper it was also pointed out that during the infantile and juvenile period the For. magn. shifts towards the occiput. It appears now that also the slope of the Foramen changes during growth. *For in the skull of a young ape the Foramen magnum lies more horizontally than in that of a full-grown one.* The following may serve as a proof of this. Whereas in a full-grown Siamanga the angle varied between 55° and 68°, I found in a juvenile skull (mixed dentition) an angle of 70°, and in an infantile skull (complete lactal dentition) an angle of 81°. In a Chimpanzee, with a complete set of milk teeth, the For. magn. lay almost horizontally with a angle of 88°. In three infantile Orang-outang skulls I found angles of 78°, 85°, and 86°, while finally a juvenile Gorilla skull had a angle of 87° and an infantile one even of 95°. In the case of this last skull, thus, the For. magn. looked forwards as in that of man. We shall soon see that as regards human beings also the plane of the For. magn. turns during infantile and juvenile periods in the same manner as with the Anthropoids, though I must here point out that this turning is much more pronounced in Anthropoids and Siamanga than in human beings.

Thus both in the position and the slope of the For. Magn. the young Anthropoid agrees more with the human conditions than the full-grown one.

In respect to the slope of the For. magn., man occupies a distinct place among all Primates, as in him the opening is not turned towards the back but towards the front. This fact, which has already been alluded to by Daubenton, and after him by all the writers on this subject, is illustrated by the figures below. I call to mind that

an angle of 90° obtained by my method agrees with a position of the For. magn. parallel to the base-line, i. e. a horizontal position.

Angle of the For. magn. in full-grown human skulls.

Papuans:	96°, 99, 99, 99, 100, 101, 103, 107, 107, 108,	Av. **101,9°.**
Negroes:	92°, 96, 07, 98, 99, 100, 100, 101, 103, 106,	„ **99,2°.**
Frisians:	86°, 89, 90, 94, 95, 99, 100, 103, 103, 103,	„ **96,2°.**
Zeelandians:	93°, 97, 99, 100, 101, 103, 104, 105, 109, 112,	„ **102,3°.**
Javanese:	92°, 92, 97, 99, 100, 100, 103, 103, 103, 105,	„ **99,4°.**

The averages of three of the groups lie comparatively near each other, and the existence of a difference between dolichocephalic skulls (the first three groups) and brachycephalic cannot be assumed on the ground of these figures, although the difference between the long dolichocephalic Frisian skulls and the short strongly brachycephalic Zeelandian skulls is very remarkable. It is also peculiar that among the Frisian skulls there were two in which the For. magn. looked slightly backwards (angles of 86° and 89°) and one where it lay exactly horizontal. This was caused by the particularly long clivus in these objects. That the degree of development of this part of the cranial base in human beings has a great influence on the slope of the For. magn. is proved by infant skulls. On an average the For. magn. in young human skulls has without exception a more considerable inclination towards the front than in full-grown ones, as will be seen from the following figures.

Angle of the For. magn. in children's skulls.

0—1	year. 110, 110, 109, 105, 104, 103, 102, 101, 100, 100, 92.
1—2	years. 100, 110, 110, 108, 106.
2	years. 107, 107, 106, 106, 103, 101, 95.
3	years. 110, 110, 108, 107.
4	years. 114, 109, 106, 105, 100.
5—6	years. 114, 113, 109, 107, 105, 103, 96, 96.
7	years. 108, 100, 100, 99, 98.
8—9	years. 104, 103, 101, 97.
10—11	years. 110, 104, 104, 101, 100 92.

The average angle of the human full-grown skulls can from the preceding table be set at 100°. And now it is seen that of the 31

skulls of children under 5 years of age only 2 have a smaller angle while of the 23 skulls of children between 5 and 12 years of age this is so in 6 cases. From this it may be inferred that during infancy when, as has been shown in the 1st paper, a shifting of the position of the occipital foramen takes place in man, also the plane of the For. magn. turns, and in the same direction as with the Anthropoids. Yet, as has been said, this turning, like the accompanying shifting of position is more marked in Anthropoids than in human beings.

We have now seen twice over that a shifting of the For. magn. and a change in the angle go hand in hand during the individual development. For in human beings as well as in Anthropoids the shifting backwards diminishes the angle of inclination. To what degree this relation between these two features exists in comparative anatomy will be apparent from the following table. The 2nd column gives the average of the angle, while the first column shows the average basal-index as determined in the 1st paper. I may here call to mind that the greater this index is, the further backwards does the For. magn. lie.

	Index basalis.		*Angle of inclination of the For. magn.*	
Lemur albifrions	87	(1)	40°	(1)
Propithecus diadema	80	(2)	42°	(2)
Mycetes	86	(3)	47.5°	(3)
Pithecia	74	(6)	58.5°	(4)
Hapale	71	(8)	65°	(8)
Cebus	67	(10)	67.8°	(11)
Ateles	64	(13)	72.7°	(16)
Chrysothrix	59	(18)	66.6°	(10)
Inuus	65	(12)	71.2°	(14)
Cynocephalus	65	(12)	64.2°	(7)
Macacus	64	(14)	73.8°	(17)
Cercopithecus	57	(19)	79.2°	(19)
Semnopithecus	74	(7)	62.8°	(6)
Colobus	75	(5)	68°	(12)
Siamanga	76	(4)	60.2°	(5)
Hylobates	71	(9)	65.1°	(9)
Chimpanzee	64	(15)	74.3°	(18)
Gorilla	61	(16)	71.8°	(15)
Orang	61	(17)	70.3°	(12)

In brackets after the figures of both series is given the place number which each of the genera would take in a regular classification. A comparison of these place numbers shows at a glance in how far the position and the slope of the For. magn. go hand in hand. In general there appears to be a decided parallelism between these features in monkeys, and only in a few cases there is a fairly marked difference between position and slope. This is, for instance, the case in Chrysothrix where the angle is small in comparison to the position, and in Colobus where the reverse is the case.

At the beginning of this paper mention was made of the opinion held by HUXLEY, viz. that the slope of the For. magn. is in proportion to the degree of prognathism. In a following communication, which will deal with the prognathism of the primate skull, this view will be discussed at greater length.

Physics. — "*A short reply to* Mr. VAN LAAR'S *remarks.*" By Prof. PH. KOHNSTAMM. (Communicated by Prof. J. D. VAN DER WAALS).

In the proceedings of the preceding meeting of this Academy Mr. VAN LAAR made some remarks suggested by a paper by Mr. TIMMERMANS and me. Though these remarks do not call in question in any point the validity of our results, but exclusively deal with the question whether we have done sufficient justice to the share Mr. VAN LAAR has had in the construction of the theory, I think that both politeness to Mr. VAN LAAR and deference to the communicator of these remarks forbid me to leave them unanswered. So I shall try to state as shortly as possible the reasons why I still think I have done full justice to that share.

1. Mr. VAN LAAR writes in point *a* of his remarks:[1]) "Here I must remark that I have *never*[2]) represented the special case $a_{12} = \sqrt{a_1 a_2}$ as the general case."

In writing this Mr. VAN LAAR had certainly forgotten that he wrote in These Proc. Sept. 1906 p. 227: "In the *third* paper in These Proceedings (June 24, 1905) the equation:

$$\Delta = \frac{1}{T_1}\left(\frac{dT}{dx}\right)_0 = \theta\sqrt{\frac{1}{\pi}}\left[\theta\sqrt{\frac{1}{\pi}}\left({}^3/_2 - {}^1/_2\sqrt{\frac{1}{\pi}}\right)^2 - 1\right] \ldots (3)$$

was derived ... for the *quite general*[2]) case $a_2 \lessgtr a_1 \quad b_2 \lessgtr b_1$", etc.

And on the same page: "Now the restricting supposition $\beta = 0$

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[1]) These Proc. XII p. 455.

[2]) Mr. VAN LAAR's italics.

was relinquished for the determination of the *double point* of the plaitpoint line, and the *quite general case*[1]) $a_2 \gtrless a_1$ $b_2 \lessgtr b_1$ was considered.

And on p. 228: "We can, namely, characterize *all possible pairs*[1]) of substances by the values of θ and π, and finally it will *only*[1]) depend *on these values*,[1]) which of the three main types will appear."

And on p. 230: "The calculations get, however, so exceedingly intricate that they proved practically unfeasible for the *general case*[1]) $a_2 \gtrless a_1$ $b_2 \lessgtr b_1$."

And on p. 231: "This appears already from the fact that the substitution of the *quite general assumption*[1]) $b_1 \lessgtr b_2$ for the simplified assumption $b_1 = b_2$ has made no change in the existence of a double point . . ., and that also the calculations for the limits of type III . . . may be carried out for the *quite general case*[1]) $b_1 \gtrless b_2$."

And on p. 232: "The calculation proves that in the *quite general case*[1]) $b_1 \lessgtr b_2$" etc.

For, everywhere where the *general case* is spoken of here, it is the case $a^2_{12} = a_1 a_2$ that is meant, and also the quotation from p. 228 is possible only, by an identification of the general case and this special one.

2. In point *b* of his remarks Mr. VAN LAAR says in connection with our sentence that his investigations: "very onesidedly, lay the stress on the existence of open plaits, a circumstance which by no means can be considered as a *result*[2]), as it immediately follows from the arbitrary, if not erroneous *supposition*[2]) of the linear dependence of *b* and *x*": "Now I have never asserted that $\frac{d^2b}{dx^2} = 0$ would always agree with what actually happens; again I have simply *assumed*[3]) this in order to make the *calculations*[3]) possible."

Yet I read on p. 231 of the cited paper: "We shall once more emphatically point out that the *numeric*[3]) results of our investigation will naturally be modified, when *b* is not assumed to be independent of *v* and *T* . . . but that *qualitatively*[3]) everything will remain unchanged."

And on p. 233: "Then further increase of pressure makes the phases 1 and 2 again diverge . . . without the longitudinal plait ever closing again — *as was formerly considered possible*[1]) — [cf.

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[1]) The italics are mine.
[2]) T. and K's italics.
[3]) Mr. VAN LAAR's italics.

inter alia VAN DER WAALS, Cont. II p. 190 (1900)]. Only at temperatures higher than T_0 . . . there can be question of homogeneity to the highest pressures."

It seems to me that every unprejudiced reader of these lines must acknowledge that Mr. VAN LAAR thought that he gave a new *result* here, materially differing from the result of a closed plait as it was thought possible by VAN DER WAALS, and that he cannot possibly have realized when writing these lines that this divergent result was only founded on his assumption $\frac{d^2b}{dx^2} = 0$.

3. As to point *c*, the sentence mentioned there really refers to a paper by Mr. VAN LAAR earlier than April 1905 (viz. of January 1905). I did not know, however, until the publication of the "Remarks", (and now I only know it from these "Remarks") that Mr. VAN LAAR has abandoned his views of this previous paper. Else we should, of course, not have mentioned it.

4. With regard to point *d* we must protect Mr. VAN LAAR against himself. We had said: "His results are of importance particularly because they showed that under certain circumstances non-miscibility can occur for perfectly normal substances, a fact which was generally doubted at the time." Mr. VAN LAAR remarks in this connection that it was by no means generally doubted up to now whether miscibility could occur for normal substances but only whether some special "abnormal" forms of non-miscibility could occur for perfectly normal substances. I must maintain in opposition to this that both LEHFELDT and VAN DER WAALS, to whom we referred l.c., had by no means a special case of non-miscibility in view, but very decidedly all non-miscibility. So Mr. VAN LAAR's merit is decidedly greater than he will own here. On the other hand I must confess that in our endeavours to be perfectly objective to Mr. VAN LAAR, we have really got unjust in the above cited sentence to Mr. VAN LAAR's predecessors: VAN DER WAALS and KORTEWEG. The above statement might lead one to think that Mr. VAN LAAR had been the first to demonstrate the possibility of non-miscibility for normal substances. As Mr. VAN LAAR justly remarks: this is incorrect, and it would have been better if our sentence had run like this: His results are of importance particularly because he adhered to the possibility of non-miscibility for normal substances in a time in which this was pretty generally doubted, and showed once more that for certain values of *a*'s and *b*'s, which could not a priori be considered as improbable, non-miscibility must really appear".

If I wanted to discuss also Mr. VAN LAAR's other remarks, I should

have to enter fully into the very heart of the matter, as I cannot assume the reader to be fully acquainted with the details of these investigations. But then I should think I abused the hospitality which this Academy so courteously extends in its publications also to non-members. So I think that the above will suffice. If Mr. VAN LAAR should, however, wish to pursue this discussion elsewhere, I am willing, though not desirous, to continue it.

Chemistry. — *"The equilibrium solid-liquid-gas in binary systems which present mixed crystals."* By Dr. H. R. KRUYT. (Communicated by Prof. P. VAN ROMBURGH.) First communication.

In the Archives Néerlandaises [2] **5** (Jubilee number in honour of Prof. LORENTZ) p. 360 (1900) Prof. BAKHUIS ROOZEBOOM published an article "Sur l'équilibre de cristaux mixtes avec la phase vapeur" in which he described and illustrated the ptx surface of a binary system when exclusively homogeneous mixed crystals occur as a solid phase. He treats the case of unlimited miscibility in all phases and especially for a system in which the melting point line proceeds without a maximum or a minimum. He has, moreover, limited himself to the case that the three-phase line solid-liquid-gas (SLG) also occurs without a maximum or a minimum.

These matters have not been further investigated theoretically [1]; there was in fact no inducement to do so, as there has been an almost entire absence of experimental research. Only two investigators, SPERANSKI [2]) and KÜSTER [3]) furnished material as to the equilibrium of mixed crystals with a gas-phase, whereas the researches of HOLLMAN [4]) belong to a category of more complicated phenomena.

I intend to carry out a series of investigations in order to extend our knowledge of the systems showing a miscibility in the solid condition. First of all, I will accept the facts already known and, therefore will discuss at present, *theoretically*, the various possibilities of the progressive change of the three-phase line indicated by ROOZEBOOM (l.c.) and communicate later the results of an *investigation*

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[1]) The results obtained by A. SMITS (Proc. (1908) XI p. 165, and Zeitschr. f. physikal. Chem. (1909) **67**, 464) do not differ from those of ROOZEBOOM. The only paper I know connected with this subject is a communication of MEYERHOFFER: "Ueber Reifkurven", Zeitschr. f. physikal. Chem. **46**, 379 (1903).

[2]) Zeitschr. f. physikal. Chem. **46**, 70 (1903) and **51**, 45 (1905).

[3]) Ibid. **51**, 222 (1905).

[4]) Ibid. **37**, 193 (1901).

as to the three-phase equilibria in the system *p*-dichlorobenzene — *p*-dibromobenzene, the same system of which, thanks to KÜSTER and SPERANSKI (l.c.), we already know a series of solid-gas equilibria.

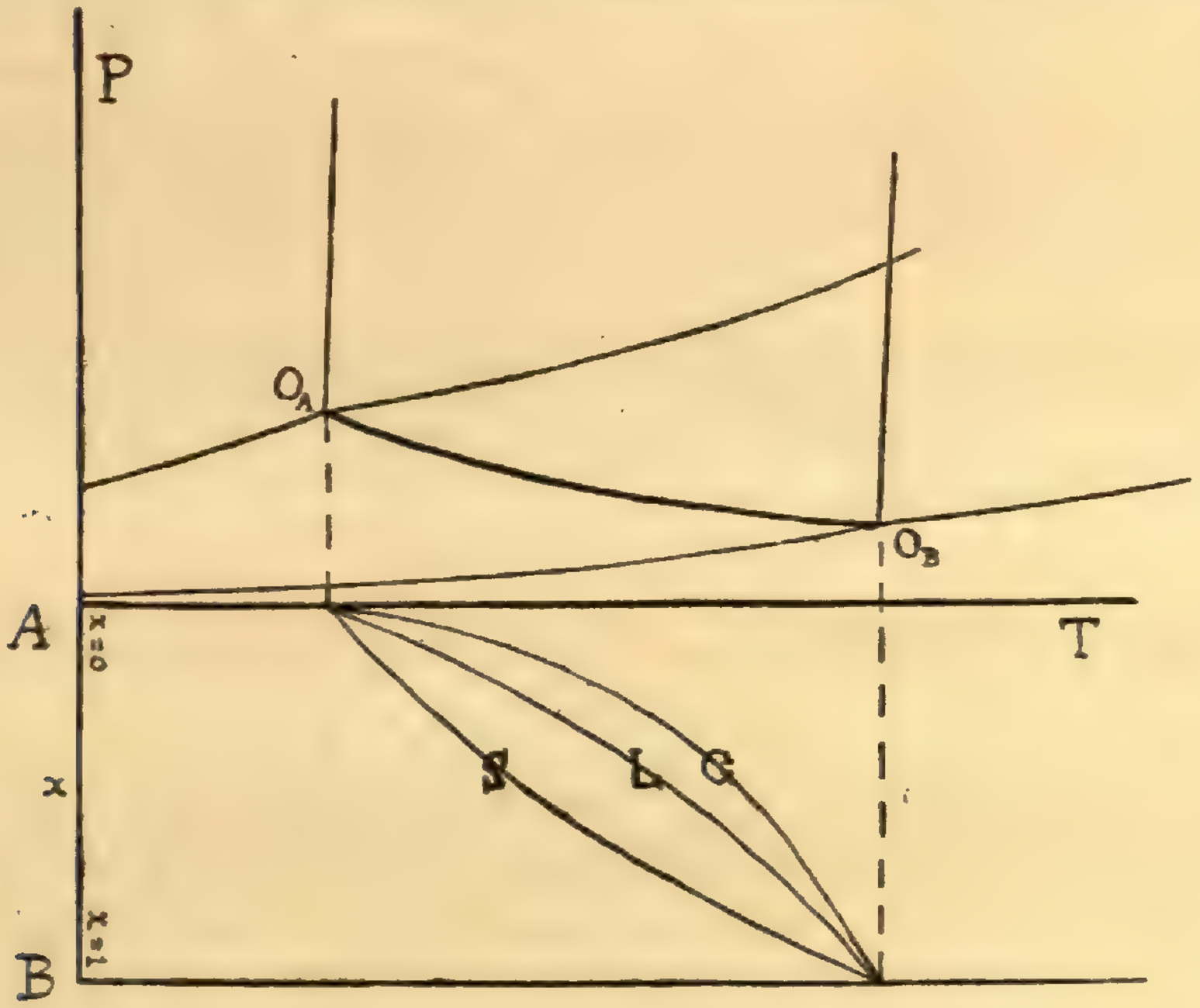

Fig. 1.

Fig. 1 is a combined PT and Tx-projection: O_A and O_B are the triple points of the components. They are connected by the three-phase line. In the Tx projection this line divides into three branches which indicate, respectively, the composition of the solid (S) liquid (L) and gas (G) phases.

Since the influence of the pressure on the equilibrium LS, is very trifling and as triple-point pressures are comparatively low, the branches S and L may, usually, be taken as being equal respectively to the melting-point curve and the freezing-point curve [2]) at 1 atmosphere.

In fig. 1 is assumed $P_{O_A} > P_{O_B}$ [1]) which case we will call chief type I. We will now ascertain under what conditions three conceivable cases might occur, namely:

case *a* with a maximum pressure in the three-phase line
„ *b* „ „ minimum „ „ „ „ „
„ *c* without a max. or min. „ „ „ „

To get an insight as to the change of the pressure with the tem-

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[1]) *A* (as is customary) is the name of the component with the lowest melting point and with a vapour pressure greater than that of *B* at the same temperature.

[2]) In what follows we shall speak of these curves "as the branches of the melting diagram."

perature we must first of all proceed in the direction indicated by Prof. VAN DER WAALS [1]) where he treats of the three-phase equilibria of a binary compound with liquid and vapour.

To the ψvx-surface of the liquid and vapour condition another one has to be added which shows the connection between those quantities in the homogeneous solid phase. If we consider the case occurring most frequently that the fusion takes place with an increase in volume this surface will be found between the liquid-vapour surface and the ψx-plane.

As to the form of this new ψvx-surface it should be observed that it will practically be a plane with descriptive lines proceeding from the ψv-plane for $x = 0$, to that for $x = 1$. For the mixing of two solid substances to a homogeneous solid phase takes place either *without* a change in volume or *with a hardly appreciable* one [2]).

If we now wish to know which are the coexisting phases we must allow tangent planes to move over these surfaces and thus cause the appearance of the derived surfaces and connodal lines [3]).

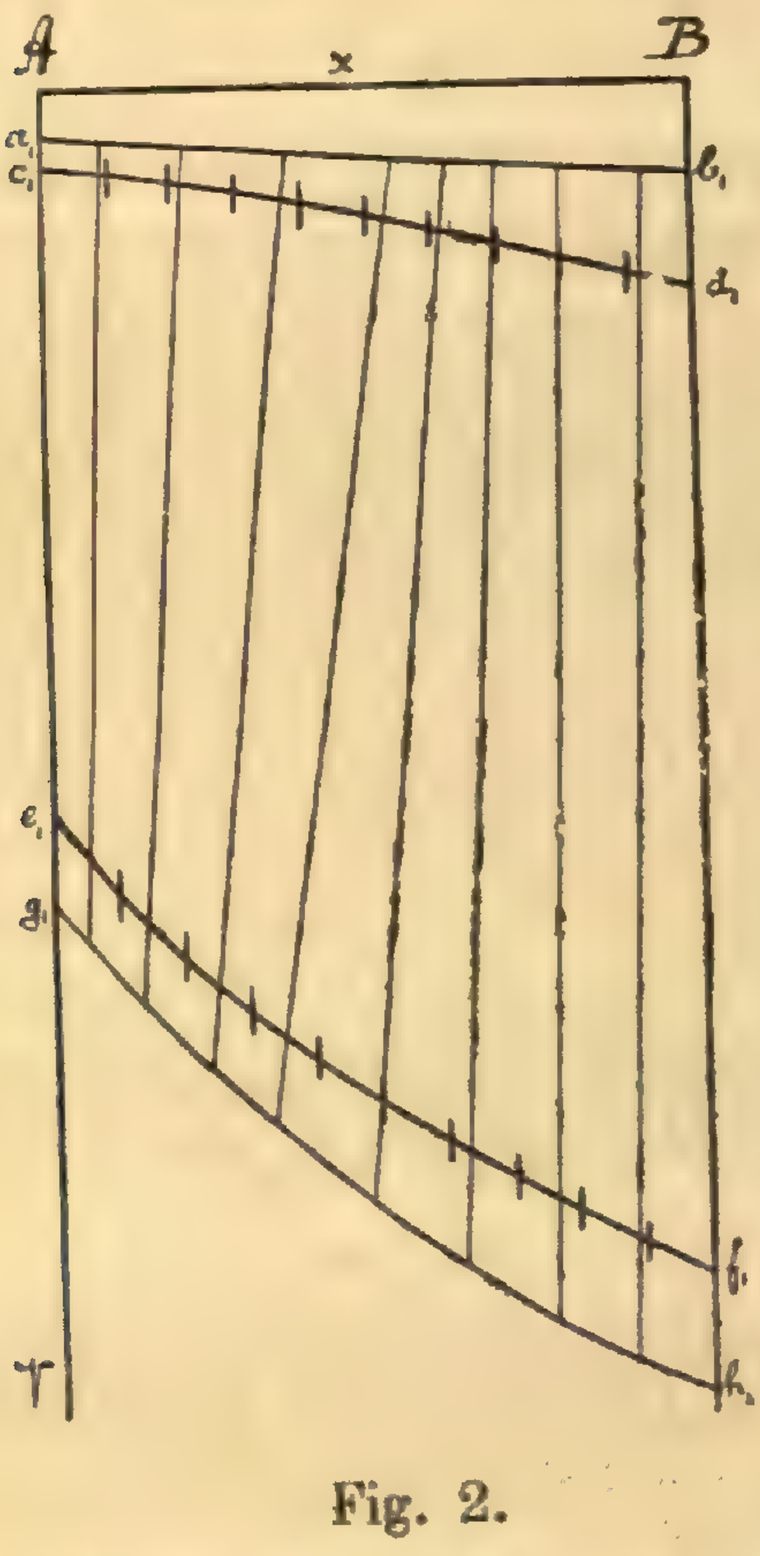

Fig. 2.

Let us commence by considering a surface for a temperature below the triple-point temperatures of the components. The surface for the solid condition will then be situated very low, the tangent plane will rest both on this surface and on the vapour part of vapour-liquid surface. The lines a_1b_1 and g_1h_1 in fig. 2 indicate the connodal lines so formed. The derived surface thus obtained will be situated lower than the derived surface which rests on the two parts of the vapour-liquid surface and which, therefore, does not represent stable conditions, but the vapour equilibria of "supercooled" liquids. The connodal lines (c_1d_1 and e_1f_1) proceeding therefrom are situated between the connodal lines of the solid-vapour equilibrium.

If we proceed to a higher temperature the correlated connodal lines

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[1]) Verslagen Kon. Akad. V, p. 482, (1897).

[2]) Cf. RETGERS, Zeitschr. f. physikal. Chem. **3**, 497 (1889) and
GOSSNER, „ „ Kristallographie **44**, 417 (1908).

[3]) In what follows, the question whether a minimum or a maximum pressure is possible for the coexistence of two phases has not been considered. All nodal lines are therefore supposed to proceed in the same sense.

approach each other; and also the stable and metastable branches on the vapour part especially at the side of the component melting at the lowest temperature [1]). For if we approach the temperature of the triple point of this component the points e_1 and g_1 of fig. 2 will have coincided to the point $e_2 g_2$ in fig. 3, which is intended for the temperature of O_A (fig. 1). The two derived surfaces intersect each other in the ψv-plane of the component A; that intersecting line is, of course, the tangent to the ψ-line for the gas-liquid condition of A and just the one which is also tangent to the ψ-line of solid A (triple point A).

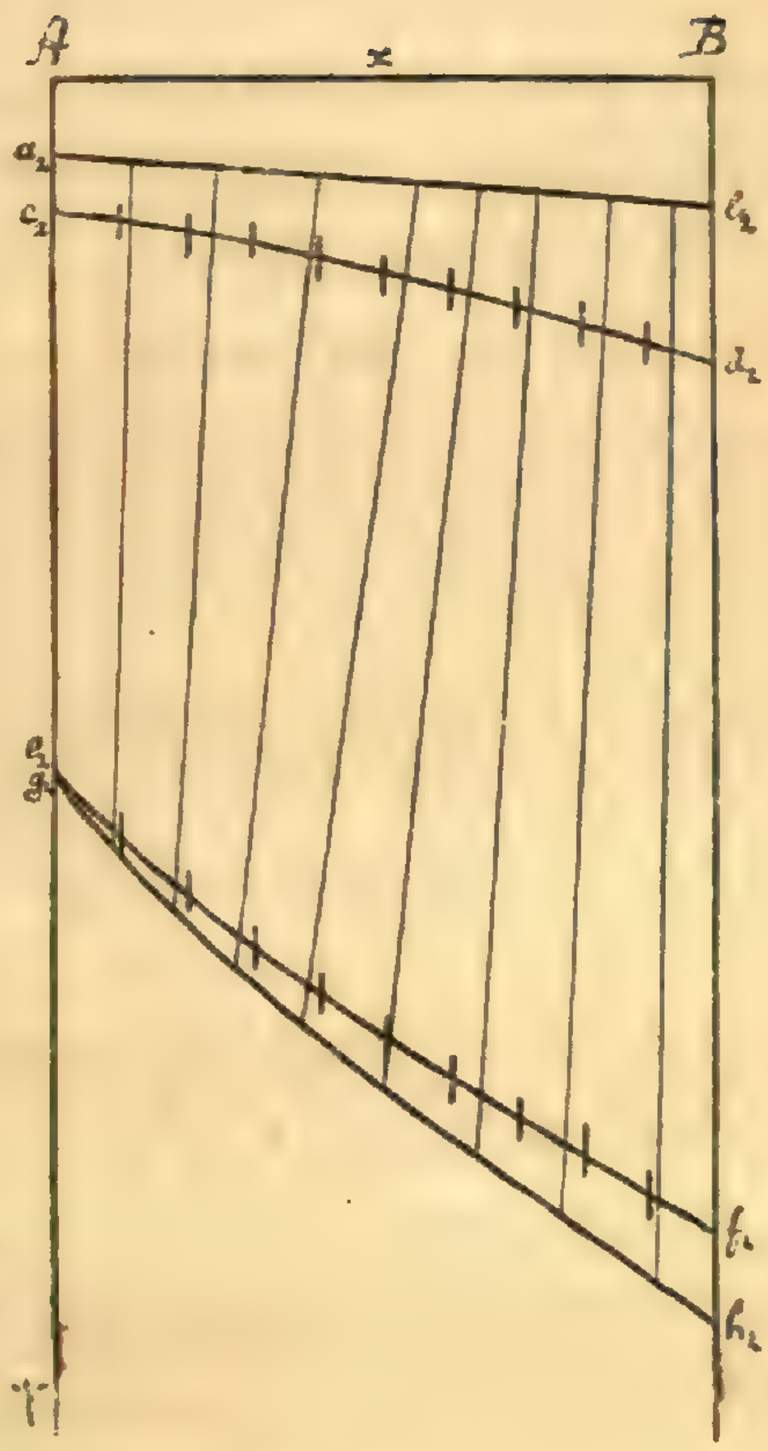

Fig. 3.

By consulting fig. 4 it will be easily seen what happens at a temperature situated between that of the two triple points. The rolling tangent plane coming from the A side will now rest first on the liquid- and vapour parts; but if a certain nodal line pq is thus reached the tangent plane will rest also on a point r of the surface of the solid phase. The angular points of the three-phase triangle pqr give us the composition of the three possible coexisting G, L, and S phases at that temperature. By further motion of the tangent plane a derived surface for GS equilibria is formed, whilst also a similar movement over the liquid part of the fluid surface and over the surface of the solid phase is possible in the direction of the small volumina. Hence a new system of connodal lines for LS equilibria is formed starting from r and q. Fig. 4, however, will be plainly understood without further comment and a discussion of the configurations at higher temperatures will also be superfluous.

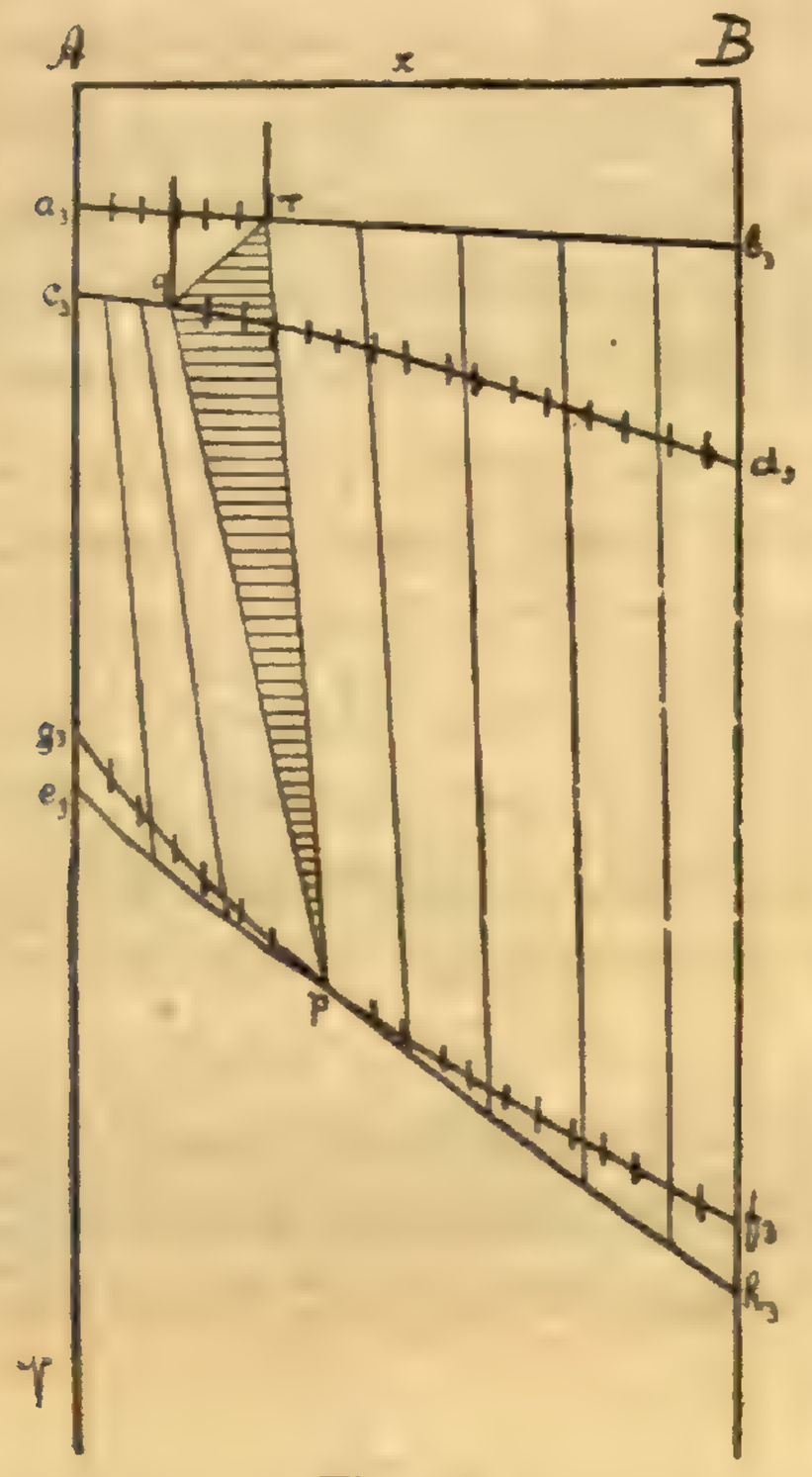

Fig. 4.

[1]) The non-related connodal lines ab (solid) and cd (liquid) diverge from each other because as a rule the coefficient of expansion of a substance is smaller in the solid than in the liquid state.

Prof. VAN DER WAALS (l. c. p. 490) has also taught us how to deduce an expression showing the relation between p,t and x.

From the three equations

$$V_S dp - \eta_S dt = dM_1\mu_1 + x_S d(M_2\mu_2 - M_1\mu_1)$$
$$V_L dp - \eta_L dt = dM_1\mu_1 + x_L d(M_2\mu_2 - M_1\mu_1)$$
$$V_G dp - \eta_G dt = dM_1\mu_1 + x_G d(M_2\mu_2 - M_1\mu_1)$$

follows

$$\frac{dp}{dt} = \frac{\begin{vmatrix} x_S & \eta_S & 1 \\ x_L & \eta_L & 1 \\ x_G & \eta_G & 1 \end{vmatrix}}{\begin{vmatrix} x_S & V_S & 1 \\ x_L & V_L & 1 \\ x_G & V_G & 1 \end{vmatrix}} = \frac{x_S(\eta_L - \eta_G) + x_L(\eta_G - \eta_S) + x_G(\eta_S - \eta_L)}{x_S(V_L - V_G) + x_L(V_G - V_S) + x_G(V_S - V_L)}.$$

This gives us a *quite general expression* for the three-phase line in the systems described. It will, however, not be easy to arrive through it to the desired elucidations. If, for instance, we wish to know when $\frac{dp}{dt}$ will be equal to 0 the numerator thus becoming nought, the question first arising is what do $\eta_L - \eta_G$ etc. really represent. KOHNSTAMM [1]) has rightly observed that such differences must not be simply called heat of condensation etc. because η_L and η_G do not relate to the same mixture. And the second question as to the numerical value of those quantities in a system to be investigated is still much more difficult to answer.

In order to get a first insight into these systems, I have taken another course though of less general applicability. We will see how the pressure changes in regard to the triple-point pressure of A, when the liquid phase has the composition x_L assuming that x_L has a very small value, in other words that but a very small quantity of B has been added to A.

The temperature T_2 at which that liquid is in equilibrium with a solid phase, the composition of which is x_S, is found from ROTHMUND's formula [2]) for very dilute mixtures:

$$T_2 = T_1 + \frac{RT_1^2}{q}(x_S - x_L) \quad \ldots\ldots \quad (1)$$

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[1]) Proc. Kon. Akad. IX p. 647 (1907).

[2]) Zeitschr. f. physikal. Chem. **24**, 710 (1897).

in which T_1 is the temperature of the triple point O_A.

The vapour pressure P_2 at the temperature T_2 is the sum of the partial pressures of the components p_A and p_B:

$$P_2 = p_A + p_B$$

for which we may write

$$P_2 = (1-x_L)\, P_{T_2} + p_B \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

if P_{T_2} represents the vapour pressure of liquid A at that temperature. If now we call P_{T_1} the vapour pressure of A at its triple point and use VAN DER WAALS' well known formula for the saturated vapour pressure we may write

$$l\frac{P_k}{P_{T_1}} = f\frac{T_k - T_1}{T_1}$$

$$l\frac{P_k}{P_{T_2}} = f\frac{T_k - T_2}{T_2}.$$

By subtraction we get:

$$l\frac{P_{T_2}}{P_{T_1}} = f\frac{T_2 - T_1}{T_1}$$

$$l\,P_{T_2} = f\frac{T_2 - T_1}{T_1} + lP_{T_1}$$

If now we substitute the value found in (1) for T_2 we obtain

$$P_{T_2} = P_{T_1}\, e^{f\frac{RT_1}{q}(x_S - x_L)}$$

thus writing (2) in this form:

$$P_2 = (1-x_L)\, P_{T_1}\, e^{f\frac{RT_1}{q}(x_S - x_L)} + p_B \quad . \quad . \quad . \quad (3)$$

If now case Ia (maximum pressure) is to occur, the three-phase line must rise from O_A to higher values of P and therefore $P_2 > P_{T_1}$. The chance of seeing this case realised in a certain system, therefore depends on P_2 having as great as possible a value in regard to P_{T_1} and relation (3) shows us when this will be the case. For the first term $\frac{T_1}{q}$ and $x_S - x_L$ will then be characteristic. The value of $x_S - x_L$ is indicated by the difference in initial direction of the branches of the melting point lines for solid and liquid and this difference is determined precisely by $\frac{T}{q}$ [1]). When therefore we pay special attention to $x_S - x_L$,

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[1]) Compare VAN LAAR, Zeitschr. f. physikal. Chem. **64**, 257 (1908).

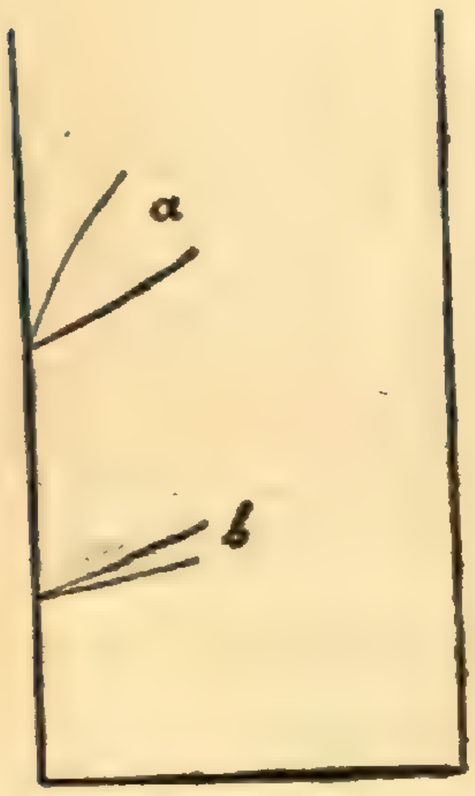

Fig. 5.

the first term of (3) will be large if $x_S - x_L$ is large, that is to say when the initial directions of the branches of the melting diagram line differ greatly (Fig. 5*a*).

The second term of (3) the partial pressure of the component B will as a rule be greater [1]) when this component gets more volatile; as in the case of this chief type I we have assumed that its triple point pressure is smaller than that of A we shall have the most advantageous conditions when they differ as little as possible.

For the case I*a* is, therefore, required 1. a type of melting diagram with greatly diverging branches near the A-axis and 2. about equal triple point pressures.

Case I*b* (minimum pressure) makes two demands: from O_A an initial fall, but followed by a rise; if this second demand is not fulfilled we are dealing with I*c*. This second demand means, of course, a small difference of the triple point pressures; the first demand, a small P_2 is, therefore, in regard to the value of p_B in (3), opposed to the second and is, in consequence, determined altogether by the first term of (3). In order that this may be as small as possible it is, of course, required that $x_S - x_L$ shall approach 0 as closely as possible, a demand which is complied with in a melting diagram with branches almost coinciding in the initial direction. (Fig. 2*b*).

We arrive at an identical result if we start from the triple point of B and examine the vapour pressure P_2' of a liquid containing a little of A, when that liquid can also coexist with a solid phase. In this case the relations (1), (2), and (3) become:

$$T'_2 = T'_1 - \frac{RT'^2_1}{q'}(x_S - x_L) \quad \ldots\ldots \quad (1bis)$$

$$P'_2 = p_A + x_L P'_{T'_2} \quad \ldots\ldots \quad (2bis)$$

$$P'_2 = P'_{T'_1}\, e^{-\frac{RT'^2_1}{q'}(x_S - x_L)} + p_A \quad \ldots \quad (3bis)$$

which will be readily understood on considering that the accentuated signs have the same significance for B as the non-accentuated ones had above for A.

In the case of I*b* the three-phase line must descend from B, therefore $P'_2 < P'_{T'_1}$. Now first of all p_A should be at a minimum

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[1]) Apart, therefore, from special differences in the critical quantities and of special influences of the components on each other.

which, on the same supposition as above, again demands about equal triple-point pressures for A and B; secondly, the exponent of e with a negative sign should be as large as possible, which requires widely diverging branches in the melting diagram at the side of the component B.

These demands put from two sides are brought into agreement by a conclusion of VAN LAAR (loc. cit. p. 265) that closely adjacent branches in the melting diagram at the side of the one component cannot meet a similar configuration at the side of the other.[1]) If this were possible, the occurrence of a maximum *and* a minimum in one three-phase line would be quite possible.

In the case of I*b* we therefore, require:

1. Melting diagram with branches nearly coinciding at the side of the A-axis and 2. about equal triple-point pressures.

Case I*c* finally occurs as an intermediate case between the two previous extreme cases. Of course, the line $O_A\,O_B$ may be concave or convex in regard to the temperature axis; this depends on whether the conditions for I*a* or I*b* have been partially fulfilled. Let us call these cases Ic_1 and Ic_2 respectively. For definite forms of the melting diagram points of inflection may probably occur, but our mode of treatment is inadequate for their investigation.

A single remark may be made as to the chance of observing a fall of the three-phase line starting from O_A. As stated, the following condition is required:

$$(1-x_L)\,P_{T_1}\,e^{f\frac{RT_1}{q}(x_S-x_L)}+p_B<P_{T_1}.$$

If now we imagine the most favourable circumstance, in which p_B may be neglected (because the components differ, for instance, very much in their melting temperature) the factor $(1-x_L)$ will cause a decrease and the factor $e^{f\frac{RT_1}{q}(x_S-x_L)}$ an increase in the value of the first member in regard to that of the second one. For $1-x_L$ is always <1; the other factor is >1 and only in the case of $x_S=x_L$ it is equal to 1: in that case a fall may be expected, but as soon as x_S and x_L differ in value the enlarging factor appears and the said difference occurs therein *exponentially*. The enlarging influence will, therefore, very soon exceed the other, so that the chance for realising the case I*c* will be diminished and that for I*b* will be reduced to a minimum.

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[1]) At least when we make the same suppositions as in the footnote on p. 543,

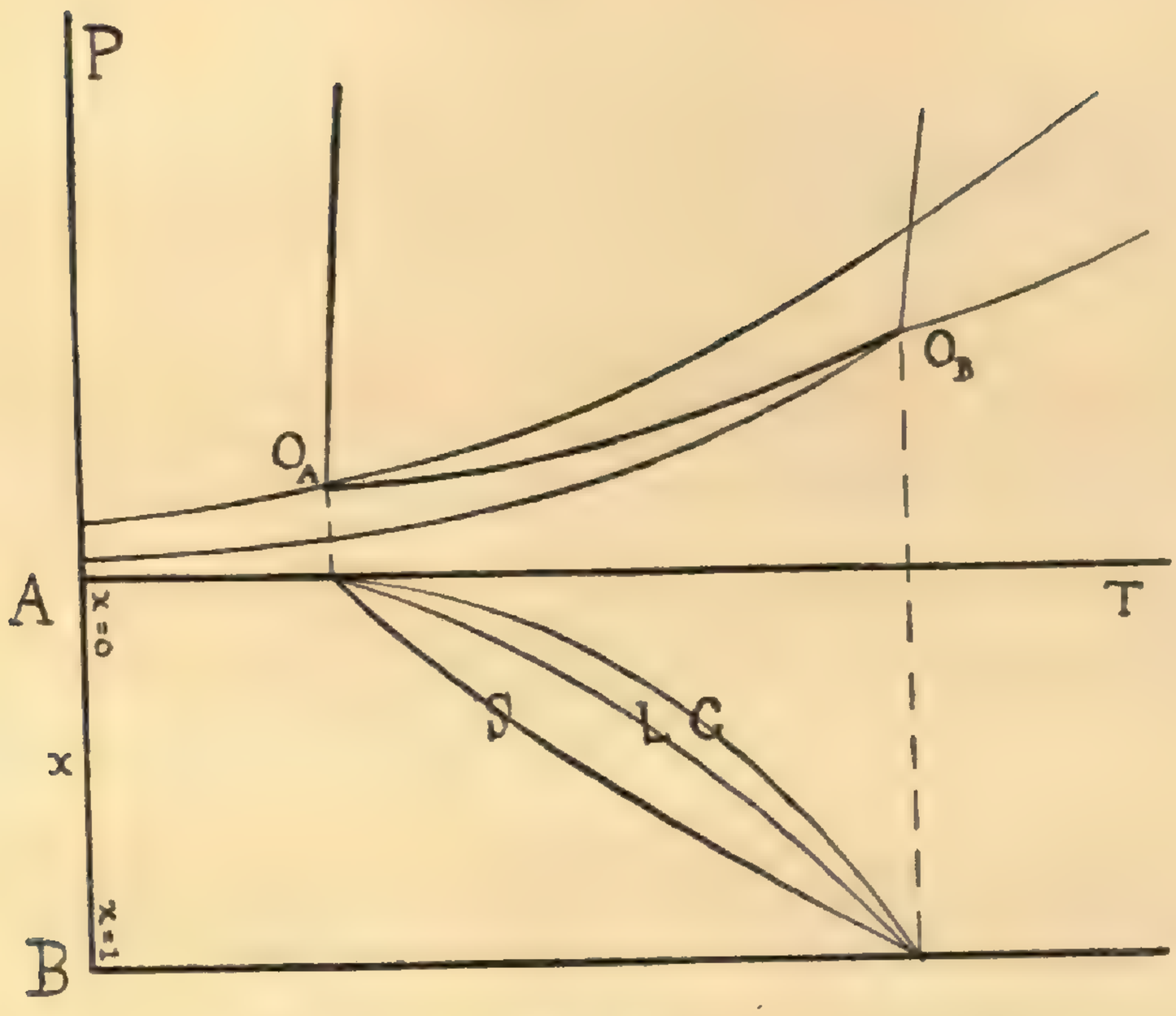

Fig. 6.

Let us now consider a second category of possibilities, namely $P_{O_A} < P_{O_B}$ which case we will call chief type II.

We again distinguish three possibilities, viz.

a. maximum pressure in the three-phase line
b. minimum " " " " "
c. no max. or min. " " " " "

It will be superfluous to repeat the previous arguments when we examine the initial directions in the equations (3) and (3*bis*). The conclusions arrived at are that we require for:

Case II*a*: a melting diagram with closely joined branches at the side of the component *B*, and but slightly differing triple-point pressures.

Case II*b*: a melting diagram with closely joined branches at the side of the component *A*, and but slightly differing triple-point pressures.

Case II*c* will be again the intermediate case between the two previous ones; a concave (IIc_1) and a convex (IIc_2) course will again be possible.

In a future paper, I hope to communicate the results of an experimental investigation of the system *p*-dichlorobenzene — *p*-dibromobenzene which has been going on already for a considerable time.

November 1909. *Utrecht*, VAN 'T HOFF-laboratory.

ERRATA.

p. 438 line 16 from the top: for 1000 read 10000.

(January 26, 1910).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETINGS
of Saturday January 29 and February 26, 1910.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van Zaterdag 29 Januari en 26 Februari 1910, Dl. XVIII).

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Geology. — *"Pienaarite, a melanocratic foyaite from Transvaal."* By H. A. BROUWER. (Communicated by Prof. G. A. F. MOLENGRAAFF.)

(Communicated in the meeting of November 27, 1909).

Among the nepheline syenites on and to the west of the farm Leeuwfontein to the north-east of Pretoria, which show a complete series of varieties in chemical and mineralogical composition, the "collection MOLENGRAAFF" contains a variety very rich in titanite, which occurs $^1/_4$ mile to the west of the Pienaarsriver near the boundary of the farm Zeekoegat.

Macroscopically the rock shows red felpars to 1 cm. up in length, which have a tabular development after (010), and smaller crystals of red nepheline, with which contrast numerous slender prisms of aegirine and bright crystals of titanite, which make up over half of the rock.

Under the microscope the rock is seen to consist of felspar, nepheline, less sodalite, much aegirine, (aegirine augite) and titanite and small quantities of apatite, fluorine, calcite, analcime, and titanic iron ore.

The felspars are orthoclase and microperthite in Carlsbad twins.

Nearly always nepheline and sodalite are transformed, respectively into pseudomorphoses of mica and zeolites. In the crystals of nepheline, which are not entirely transformed into mica, the transformation begins along the fissures, but nearly all the crystals are entirely altered. The sodalite pseudomorphoses consist of zeolites, in which we find distributed some small flakes of mica.

The aegirine is strongly pleochroic from olive-green to yellowish green, some crystals are homogeneous, other ones contain a centre of aegirine augite, which has for the greater part very low extinction angles; they are very rich in inclusions of small crystals of titanite and apatite, and they are strongly impregnated with fluorspar.

The titanite forms the well known twins after (001), in the rhombic sections the long diagonal is the twinning plane; both individuals are polysynthetically twinned. They are pleochroic from salmon coloured to colourless.

The apatite is the first product of crystallization, it is even formed as small idiomorphic inclusions in the titanite, for the greater part the crystallization of the other elements was simultaneous; the aegirine is idiomorphic in relation to felspars and felspatoids but in general the contactlines are irregular and show simultaneous crystallization. The felspar includes some idiomorphic crystals of nepheline and sodalite, mainly it is the latest product of crystallization. Probably in pneumatolytical way, fluorine, calcite, and analcime crystallized in the remaining cavities.

It is evident how much the mineralogical composition of this rock differs from that of the normal types of nepheline syenite by its high content of aegirine and titanite. A. LACROIX [1]) gave the name of covite to the mesocratic form of this group and of teralite to the melanocratic form; as the type of covite he considers the rock of Magnet Cove in Arkansas, described by WASHINGTON and as type of teralite the alkali felspar-nepheline rocks from the Crazy Mountains in Montana.

The chemical composition of the rock here described is shown in I of the following table (analysed by F. PISANI); it is compared with the analyses of some covites and teralites.

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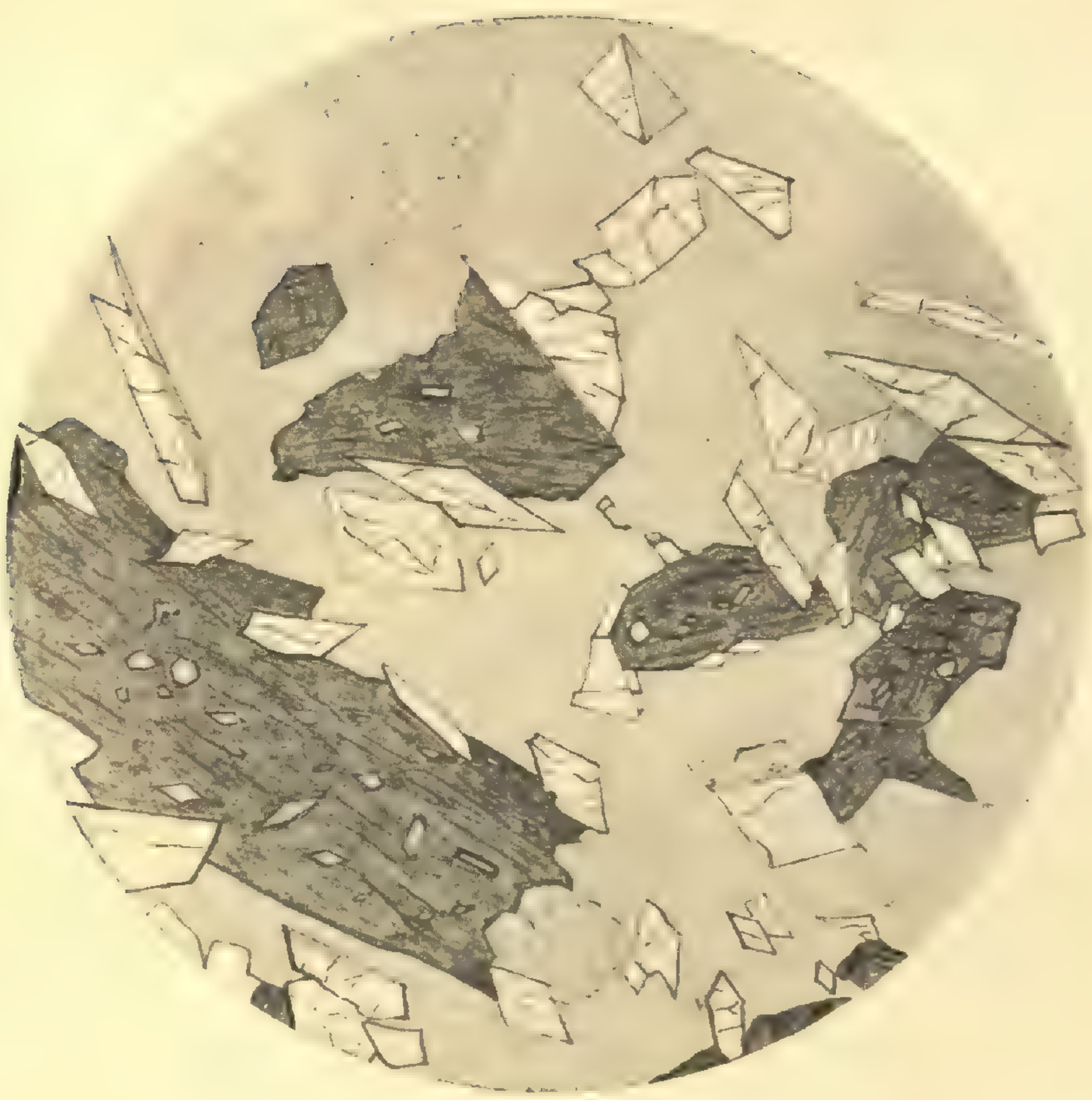
1) Matériaux pour la Minéralogie de Madagascar. Extr. nouv. Arch. du Museum. 4e serie, Tome 1, pag. 184.



Explanation of Figure.

(× 30).

A part of a large individual of felspar shows poikilitic relation to aegirine, titanite, nepheline (at the top to the left) and sodalite (at the lower edge, in the middle and to the right).

The aegirine contains numerous idiomorphic inclusions of titanite, apatite and fluorspar

	I	II	III	IV	V	VI
SiO_2	49.20	49.70	51.10	47.67	44.65	47.85
TiO_2	7.13	1.33	1.38	—	0.95	—
Al_2O_3	9.23	18.45	21.10	18.22	13.87	13.24
Fe_2O_3	7.73	3.39	0.90	3.65	6.06	2.74
FeO	3.24	4.32	5.58	3.85	2.94	2.65
MnO	—	—	—	0.28	0.17	—
CaO	11.55	7.91	5.35	8.03	9.57	14.36
MgO	1.35	2.32	2.81	6.35	5.15	5.68
Na_2O	6.20	5.33	6.35	4.93	5.67	3.72
K_2O	1.96	4.95	4.21	3.82	4.49	5.25
P_2O_5	0.06	0.40	—	2.97	2.10	2.74
H_2O	2.20	1 34	0.87		1.50	2.42
Som	99.85	99.44	99.65	100.15	99.93	100.65

I. Pienaarite. Leeuwfontein (320) Pretoria. Transvaal.

II. Covite. Magnet Cove. Arkansas cf. H. S. Washington. Journ. of Geol. IX. 614. 1901.

III. Covite. Nosy Komba. cf. A. Lacroix Mat. Minéral. Madagascar Extr. Nouv. Arch. du Museum 4e Ser. I. 32.

IV. Teralite. Crazy Mountains Bull. U. S. Geol. Surv. no. 150.

V. Teralite. „ „ „ „ „ „ „ „ „

VI. Teralite. (nepheline pyroxene malignite) cf. A. C. Lawson Bull. Dep. of Geol. Univ. of California I. 337. 1896.

We see how the rock, here described, differs in mineralogical and chemical composition from the melanocratic nepheline syenites, which are hitherto known, its characteristic features are the large amount of Fe_2O_3, TiO_2, and CaO (abundance of aegirine, aegirine-augite and titanite) and its low content of lime (diminuation of the felspars and felspatoids). Prof. Molengraaff proposed to me the name Pienaarite, after the Pienaarsriver because the locality, where he collected this rock, is situated in a region between a tributary of the Pienaarsriver called Mundtspruit, and the above river itself.

Physiology. — Prof. PEKELHARING offers a communication, also in the name of Dr. C. J. C. VAN HOOGENHUYZE: "*About the formation of creatine in the muscles at the tonus and at the development of rigidity.*"

(Communicated in the meeting of December 24, 1909).

On a preceding occasion [1]) in this Academy I have made a communication concerning an investigation by Mr. VAN HOOGENHUYZE and Mr. VERPLOEGH about the excretion of creatinine in man, from which it, appeared that the excretion of this substance in a sufficiently nourished person was not increased by muscular labour. Since that time the investigation, which promised new results, now that a good method for the determination of the creatinine had been furnished by FOLIN, has been continued by VAN HOOGENHUYZE and VERPLOEGH and by several others. From these investigations has arisen the opinion that, through the consumption of protein in the tissues of vertebrate animals, creatine is formed, and that this matter is partly decomposed under oxidation, partly, particularly by the liver, changed into the anhydride, creatinine, that further the creatinine thus formed is for the greater part removed from the body through the kidneys [2]).

If this opinion is correct, one cannot but assume that the creatinine excreted by the kidneys, originates for the greater part from the creatine of the muscles, not only because the muscles are richer in creatine than other organs, but also because it is especially the muscles which contain so considerable a part of the proteins which the body contains. This supposition did not seem unacceptable, notwithstanding it has been found that the excretion of creatinine is not increased by muscular labour. Observations were made which pointed to a connection between the excretion of creatinine and another phenomenon, which has to be distinguished from the muscular contraction in a narrower sense, the muscular tonus. VAN HOOGENHUYZE and VERPLOEGH found the excretion during the night to be smaller than in the daytime; likewise did they find remarkably little creatinine in the urine of old men and of patients who had a number of muscles paralyzed, whereas in case of fever the urine appeared to contain more creatinine than usual.

That really the tonic shortening of the muscle is brought about in another way than in case of rapid contraction, which has been examined so much more, and that, when a single stimulus is followed

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[1]) Proceedings of the meeting of 30 Sept. 1905.

[2]) See: Zentrallbl. f. d. ges. Physiol. und. Pathol. d. Stoffwechsels. 1909. No. 8.

by a slow contraction, the two ways collaborate, is rendered very probable.

Already more than 20 years ago GRÜTZNER [1]) made the supposition that the long continued contraction was brought about by another kind of muscular fibres than those which cause a more rapid contraction. In the first case fibres of the type of the red, in the second fibres of the type of the white muscles, were supposed to be brought into play. Afterwards others, more particularly BOTTAZZI [2]), have defended the theory that the double contraction is caused by two different component parts of the same muscular fibre, the rapid by the double-refracting fibrils, the slow ones by the sarcoplasma. Mosso [3]) had objections against this theory and drew attention to the double innervation of the muscular fibres, not long ago once more made clear in this Academy by BOEKE [4]).

Meanwhile, whatever the opinion may be, at any rate there is some reason to assume that the contractions of two kinds must be accompanied by a chemical action of two kinds. Now that in the usual muscular labour, which is principally based on rapid and tetanic contractions, no increase in the consumption of protein and in the excretion of creatinine was found, it might be asked, whether perhaps in the tonic contraction formation of creatine in the muscles could be proved.

That under certain definite circumstances the muscles at their contraction yield more creatine to the blood than otherwise, has already been found by WEBER [5]), with respect to the heart treated after LANGENDORFF's method and beating in RINGER's solution. He also found a considerable increase in the excretion of creatine with a dog, after violent cramps had been caused in the animal with cinchonine. Not only in the last case, in which the animal for an hour "was in violent tonic and clonic cramps", but also with respect to the heart taken from the body, it may be assumed that tonus has played a part.

However, in order to draw more certain inferences, we have examined the quantity of creatine in muscles, under circumstances which, as much as possible, allowed to judge about the influence, either of tonic, or of rapid contractions.

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1) PFLÜGER's Archiv. Bd. XLI, S. 280.

2) Journ. of Physiol. Vol. XXI, p. 1, Arch. f. Physiol. 1901, S. 377, Arch. Ital. de Biol. T. XLII, p. 169.

3) Arch. Ital. de Biol. T. XLI, p. 183.

4) Proceedings of the Meeting of 23 April 1909.

5) Arch. f. exp. Path. und Pharm. Bd. LVIII, S. 93.

The determinations always took place in the same way. The minced muscles were for some hours at a stretch boiled in 0.1% HCl, in consequence of which the tissue breaks altogether and all the creatine passes into the liquid. By evaporation the extract after being freed from protein, was concentrated and then with then double volume of normal HCl heated to 115° C. in the autoclave for half an hour, by which the creatine is completely changed into creatinine. Then the determination took place, according to the method of FOLIN, with the colorimeter formerly shown in the meeting of this Academy.

First of all we have some observations to mention about decrease of the quantity of creatine in muscles, of which the tonus, in consequence of section of the nerve, was eliminated. In the outset we came to very irregular results when examining the muscles of the hindlegs of rabbits and of a young dog, after a one-sided cutting of the nervus ischiadicus. Now less creatine was found in the muscles of the paralyzed leg, now more creatine in those of the paralyzed side. The cause of this irregularity appeared to be that we did not compare exactly corresponding muscles. Especially in the rabbit the difference in quantity of creatine in white and red muscles is rather considerable. With 10 rabbits we found in the gastrocnemius on an average 4.463, in red muscles (soleus, semitendinosus and semimembranosus examined together) 2.925 mgr. creatinine in 1 grm. of the muscle.

When this source of error was avoided, the influence of section of the ischiadicus became clear. With 3 rabbits we have, taking into consideration what has been said above, repeated the experiment and three days after the section of the nervus ischiadicus, we have examined the gastrocnemius of the paralyzed and of the not paralyzed leg. We found:

	Not paralyzed.	Paralyzed.	Loss after paralyzing.
I	4.703	4.232	0.471 mgr. cr. p. gr. of the muscle
II	4.448	4.289	0.159 " " " " " " " "
III	4.983	4.013	0.970 " " " " " " " "

Although the differences found lie without any doubt beyond the limits of the errors of observation, yet we have not continued these experiments, because too little can with certainty be concluded from them concerning the influence of the tonus. It is true, the muscles of the leg the ischiadicus of which has been cut through, are distinguished from those of the other side by the loss of tonus, but there are also other differences, which are perhaps of importance, as WEBER has already observed, who also found the quantity of creatine in

the muscles of the paralyzed leg smaller than in those of the normal leg in a dog after section of the ischiadicus. That the normal leg continues performing voluntary movements, is no objection, there being no ground to assume that then creatine is formed. But the muscles of the paralyzed leg degenerate. Although we killed the animals already three days after the section of the nerve, yet every time the paralyzed gastrocnemius appeared to be of a smaller weight than the normal one. Nothing is known about the formation and the destruction of creatine in degenerating muscles. Of no less importance seems to be the change in the circulation of the blood after section of the nerve, in consequence of which the removal of creatine from the muscles may be altered in a quite incalculable degree.

On account of these objections we thought of entirely giving up the attempt to inquire into the influence of the muscletonus in warm-blooded animals, and of being obliged to occupy ourselves only with cold-blooded vertebrates, in which without any trouble the blood-circulation can be shut out (in muscles of invertebrates no creatine has been found; they were not fit for our purpose accordingly) when my colleague Prof. R. MAGNUS drew our attention to a means of bringing muscles of one half of the body of a cat into strong tonus, whilst the corresponding muscles on the other side, without any disturbance in the action of the centrifugal nerves and in the circulation of the blood, remain slack.

SHERRINGTON [1]) has found that, when in deeply narcotised dogs, monkeys, cats, rabbits or cavias, the action of the cerebral hemispheres is excluded by a section in the region of the hindmost corpora quadrigemina, after a short time the so-called "decerebrate rigidity" develops itself, a long continuing tonic contraction of definite muscle-groups, among which especially the extensors of the extremities and the retractors of the head and the neck are the chief. This state of things is dependent on impulses which arise in the periphery, and by centripetal nerves are led to the spinal cord. That is why the stiffness does not arise in those parts of which the corresponding dorsal roots are severed.

Now Prof. MAGNUS had the kindness to operate upon five cats in such a way that one foreleg was brought in tonus two, three hours at a stretch, whilst the other leg remained slack. The perfectly narcotised animal, the narcosis being brought about first by means of ether, then by means of chloroform, after section on the left side of the hindmost roots of the four or five lowest cervical nerves and of the two highest thoracic nerves was decerebrated. Soon, the right

1) Journ. of Physiol. Vol. XXII, p. 319.

leg got into tonus, the left one remaining slack. Sometimes, in order to strengthen the tonus in the foreleg, also the spinal cord, at about the eleventh breast-vertebra, was cut through. When the tonus had lasted a few hours, the animal was killed by suffocation. Directly after the triceps brachii on both sides was prepared, minced and put into hydrochloric acid.

In all experiments we found the muscle that had been in tonus, richer in creatine than the one that had remained slack, and that, expressed in mgr. creatinine on 1 grm. muscle, as follows:

	Tonus	Slack	Difference
I	3.690	3.090	0.600
II	4.340	3.848	0.492
III	4.291	3.902	0.317
IV	3.806	3.185	0.621
V	3.198	2.963	0.235

It is remarkable that this difference pretty well keeps pace with the difference that in the experiment of the contraction of the muscles, right and left, was observed. In experiment I and still more in IV, the stiffness on the right was very beautifully developed, in II the tonus was strong, but of a shorter duration, in III the tonus on the right was good, but also the left foreleg occasionally showed some stiffness, which also occurred in V, though in a smaller degree, whilst the stiffness developed here slowly and to not so high a degree as otherwise.

We think we are entitled to derive from these experiments that by the muscles in tonus more creatine is formed than by those which are slackened. For the supposition that the difference may be attributed to an increased decomposition of creatine in the slackened muscles, it seems that there is not a single ground to be adduced.

Besides this we have made a number of experiments with frogs (Rana esculenta).

In the first place the influence of irritation with induction-currents on the quantity of creatine in muscles was examined. About this communications have been made by MELLANBY [1]) and by GRAHAM BROWN and CATHCART [2]). By direct irritation MELLANBY brought the muscles in tetanus and then he found so slight an increase of the quantity of creatine that he came to the result: "that the perfor-

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[1]) Journ. of Physiol. Vol. XXXVI, p. 447.

[2]) Bio-Chemical Journ. Vol. IV, p. 420.

mance of muscular work leaves creatine unaffected". BROWN and CATHCART stimulated the muscles by means of the nerve and found a somewhat more considerable increase, of 7% à 13%, in four experiments, at which the circulation of the blood had been excluded. If the circulation was intact, they found a little diminution, not only with frogs, but also with rabbits, where it ought, however, to be taken into consideration that, in consequence of the stimulus, the muscle was more amply provided with blood, so that an exact comparison with resting muscles is scarcely possible.

We have made experiments with frogs, in three different ways, always excluding the current of blood. First, after destroying brain and spinal cord and after section of the heart, the nervus ischiadicus on one side, laid bare high in the thigh, was cut through and stimulated with a series of rapidly succeeding induction-strokes. Each stimulation lasted $^1/_2$ minute, after which $^1/_2$ minute of rest was afforded, during about one hour. In the second place the experiment was for the rest made in the same way, but the nerve was, for about half an honr, with the help of ENGELMANN'S rhythmic polyrheotome, stimulated 24 times per minute, alternately with a closing and an opening induction-stroke. At last two more experiments were made thus: the frog was cut through transversely in the lumbar region, after which the skin of the hind part was taken away. Now this was put astride on the partition of two basins of celluloid, standing against each other and filled with RINGER'S solution, so that each leg was immersed into the liquid to about half way up the thigh. Then the ischiadicus was on one side, from the pelvis, stimulated for half an hour, 24 times a minute, with single closing and opening induction-strokes.

The quantity of creatine, expressed in mgr. creatinine per 1 grm. of muscle, was found as follows:

		Stimulated	Rest	Difference
B	I	3.490	3.418	+ 0.072
	II	3.537	3.457	+ 0.080
	III	3.629	3.550	+ 0.079
	IV	3.567	3.560	+ 0.007

		Stimulated	Rest	Difference
A	I	3.366	3.386	— 0.020
	II	3.616	3.683	— 0.067
	III	3.796	3.856	— 0.060

C	I	3.203	3.230	— 0.027
	II	3.585	3.593	— 0.008

The differences are slight and do not fall, or fall scarcely, beyond the boundaries of the inevitable errors of observation. Moreover the difference is now in favour of the stimulated, now of the not-stimulated muscles. Even if one wishes to attach some importance to the greatest

differences found in these experiments, one need not yet derive from them that the muscle during the rapid contraction forms or loses creatine. For on the one hand the decomposition of creatine in the muscle is no doubt subject to quite unknown, but certainly varying influences, whilst on the other hand long continued irritating can also give rise to some lasting contraction, tonus.

That in the frog during the muscle-tonus in contradistinction to the rapid contractions, the quantity of creatine in the muscles increases, whilst, in default of tonus, it decreases, appears from the following experiments.

In the first place the influence of the elimination of the tonus, by excluding the innervation, was examined, first while the current of blood was stopped, then with undisturbed circulation of the blood. In the former case the ischiadicus was on one side cut through and then an both sides the root of the thigh was so well tied up with an elastic ligature that the blood in the vessels of the webs stood still, in such a way that the ligature ran below the ischiadicus not cut through. In the latter case the ischiadicus was simply cut through. Three days after the section of the nerve the animals were killed and the muscles of the hindlegs examined.

		intact	cut	difference
Current of blood stopped	I	2.204	2.137	0.067
	II	2.678	2.282	0.396
	III	2.990	2.790	0.200
	IV	2.987	2.887	0.100
	V	2.726	2.551	0.175
	VI	2.833	2.688	0.145

		Intact	cut	difference
Current of blood undisturbed	I	3.784	3.342	0.442
	II	4.000	3.653	0.347
	III	4.146	3.688	0.458
	IV	3.490	3.192	0.298
	V	3.434	3.131	0.303
	VI	3.685	3.334	0.351
	VII	3.157	2.900	0.257

Without any exception, therefore, there was found in the muscles that had lost the tonus for three days, less creatine than in the unhurt leg. If the current of blood was stopped, the difference was smaller than when it went on undisturbed. Yet in the tied up legs the quantity of creatine was, also on the side of the unhurt nerve, smaller than it is usually found in the frog. It is therefore probable that on both sides, after the current of blood had been stopped, creatine was decomposed. With respect to the experiments with undisturbed circulation of the blood the same objection may be raised which has been made mention of concerning similar experiments with the rabbit, viz. that it is unknown how far, perhaps

by a change of the current of blood, the removal of creatine from the muscles is altered. It is, however, not to be assumed that the differences observed should be attributed to this.

With much more certainty, however, the connection between tonic contraction and formation of creatine in the frog may, in our opinion, be derived from another series of experiments in which, with exclusion of the current of blood, muscle-tonus was caused. We have exposed the muscles to the action of substances of quite different nature, which, however, resemble each other in the fact that they cause tonus, viz.: veratrine, nicotine, calciumchloride rhodan-natrium and coffeine.

It is especially BOTTAZZI who has pointed out the tonicizing action of veratrine[1]). If the gastrocnemius of a frog is immersed in RINGER's solution containing 1 : 20000, or even less, veratrine, stimulation of the ischiadicus with a single induction-stroke causes a contraction which lasts much longer than with a muscle immersed in pure RINGER's solution. To the rapid a slow contraction is added.

In order to examine the influence on the formation of creatine the hindlegs of a frog were brought into the above mentioned celluloid basins, of which one was filled with RINGER's solution, the other with the same solution, in which a definite quantity of veratrine had been dissolved. Now the two ischiadici were, from the pelvis, during half an hour, stimulated 24 times per minute alternately with a closing and an opening induction-stroke. After that the muscles were prepared off, and with the liquid in which they had been immersed, treated in the usual way for the determination of creatine. The result was:

	RINGER's solution	Veratrine.	Difference.
		(1 : 40000)	
I	3.442	3.561	0.119
		(1 : 20000)	
II	3.189	3.389	0.200
		(1 : 5000)	
III	3.056	3.430	0.374
IV	3.250	3.670	0.420
		(1 : 1000)	
V	3.029	3.429	0.400

In III, IV and V the legs immersed in veratrine ceased to contract before the half hour had elapsed. Besides these legs showed in the end some stiffness in these experiments.

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[1]) loc. cit.

The faculty of nicotine to cause tonic contraction of muscles, has been amply studied by LANGLEY in his experiments on receptive substances [1]). The forelegs of the frog, the flexors of which are so easy to bring in tonus, also by dripping with nicotine, would have been very fit for our purpose, if not the mass of the available muscles was so small that for a single determination of creatine a large number of frogs would be necessary. The experiments of LANGLEY, however, made us surmise that also the hindlegs would be fit for our purpose, which surmise was corroborated by the result.

First an experiment was made as follows:

After destroying brain and spinal cord 1 CC of a 1 $^0/_0$-solution of nicotine in RINGER's solution was injected into the abdomen, after which the tonic contraction of the forelegs soon made itself manifest. Half an hour after the injection the current of blood, by section of the heart, was brought to a standstill. Now the left ischiadicus was laid bare in the upper part of the thigh, cut through and for half an hour stimulated 24 times per minute with induction-strokes. Till the end the muscles reacted upon the stimulation of the nerve and at last a slight rigour was to be observed.

The stimulated muscles produced 3.491 mgr. of creatinine per grm. of muscle, the non-stimulated 3.090 mgr. Difference 0.401.

Then the experiments were made in the same way as with veratrine, with the following result:

	RINGER's sol.	Nicotine. (1 : : 100)	Difference.
I	3.286	3.766	0.480
II	3.090	3.492	0.402
		(1 : : 200)	
III	3.276	3.538	0.262
		(1 : 100)	
IV	3.037	3.401	0.364

At the end of the experiment the leg immersed in nicotine did not visibly contract any more and each time this leg was somewhat stiff.

Through an examination of the action of kalium- and calcium-salts also GUENTHER [2]) has come to the result that the muscular fibre possesses contractile substances of two kinds, one of which is made more susceptible to stimulation by K, the other by Ca.

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[1]) Journ. of Physiol. Vol. XXXIII, p. 374, Vol. XXXVI, p. 347, Vol. XXXVII, p. 165, p. 285, Vol. XXXIX, p. 235. Proc. Royal Soc. B. Vol. LXXVIII, p. 170

[2]) Amer. Journ. of Physiol. Vol. XIV, p. 73.

"The first contractile substance of the sartorius", he says, "responds quickly with a contraction when subjected to a 1 percent solution of potassium chloride. Calcium chloride in a 1 percent solution produces no contraction of the first contractile element of the sartorius, gives rise to a slow contraction of the second contractile element, and produces quite a vigorous contraction of heart muscle."

We had, therefore, to expect that excitation of muscles immersed in calciumchloride would make the quantity of creatine increase. Indeed this appeared to be the case. One basin was now filled with RINGER'S solution, the other with a solution of $CaCl_2$ isotonic with it. For the rest the experiments were made in quite the same way as the preceding one. The results are the following:

	RINGER'S sol.	$CaCl_2$ 0.72 %	Difference
I	3.177	3.820	0.643
II	3.193	3.703	0.510
III	3.340	3.894	0.554
IV	3.040	3.647	0.607
V	3.156	3.501	0.345

In the first four experiments the contraction of the muscles immersed in $CaCl_2$ left off before the half hour was past and these muscles showed distinct stiffness. In V the contractions of the muscles immersed in $CaCl_2$ were at the end of the experiment clearly to be observed and stiffness was not to be perceived.

To the examination of the action of rhodane and coffeine we were led by a communication of VON FÜRTH and SCHWARZ [1]), from which it appeared that these substances, like e.g. veratrine, are able to considerably increase the labouring faculty of the muscles. The supposition that also here the tonus, the "innere Unterstützung", of which GRÜTZNER spoke, was playing a part, we found corroborated. The two gastrocnemii of the same frog were hung, one in a vessel with RINGER'S solution, the other in a vessel with the same liquid in which some citras coffeini was dissolved, or of which the sodium chloride had been replaced by rhodan-natrium. After both muscles, under the same tension, had been fastened to registrating levers, they were now and then, by means of the nervi ischiadici laid upon a single couple of electrodes, excited with an induction-stroke. Now while the muscle immersed in RINGER'S solution after each contraction returned to its former length, or was even somewhat lengthened,

[1]) PFLÜGER'S Archiv, CXXIX, S. 525.

the muscle brought in contact with coffeine or with rhodane, whilst it continued reacting well upon the excitation, became gradually not inconsiderably shorter.

The influence upon the quantity of creatine was examined in the usual way. The following figures were found:

	Ringers' sol.	NaCNS 0.614 %	Difference
I	2.822	3.098	0.276
II	3.106	3.354	0.248
III	3.051	3.537	0.486
IV	3.129	3.459	0.330
V	2.916	3.146	0.230

Towards the end of the experiment the muscles did not contract any more. Stiffening was not to be perceived.

	Ringer's liq.	Citr. Coff.		Difference
I	3.017	3.432	1 : 100	0.415
II	3.055	3.623	1 : 200	0.568
III	3.090	3.422	1 : 400	0.332
IV	3.194	3.551	1 : 400	0.357
V	3.316	3.519	1 : 800	0.203

The leg brought in contact with coffeine was in I quite stiff after 10 minutes, in II after a quarter of an hour, the contractions leaving off. In III, IV and V the contractions remained visible till the end of the experiment. Also in those cases the rigour was clear, though in V not so strongly as in III and IV.

In all cases, without any exception, therefore, the quantity of creatine was found to be increased in the muscles that had been in tonus. The difference, except with coffeine, may even be estimated somewhat higher than the figures given, because, leaving the mentioned exception out of consideration, the tonus appeared to be accompanied by a slight increase of the quantity of water. This difference is, however, so insignificant that it need not be taken into consideration.

Increase of the quantity of creatine was found only then when the muscles had been brought into tonus by excitation. Immersion of the legs, during half an hour, in the solutions, without excitation, had no influence upon the quantity of creatine. The following experiments were made in the usual way, only with this difference, that the nerves were not excited.

Ringer's sol.	Veratr. 1 : 5000	Difference
3.954	3.954	0
	Nicotine 1 : 100	
3.544	3.510	0.034
	$CaCl_2$ 0.72%	
3.399	3.394	0.005
	NaNCS	
3.340	3.336	0.004
	Coffeine 1 : 100	
3.295	3.327	0.032

In none of these cases was anything to be perceived of stiffness of the muscles.

Therefore our results are perfectly in keeping with the opinion that the muscular fibre, when reacting upon a stimulus with a rapid contraction, works in quite another way than when it is brought in tonic contraction. In the first case it consumes non-nitrogenous matter, in the second it forms creatine, consequently consumes protein. Against the supposition of Grützner that each of these actions should belong to a special kind of muscular fibres, tells among others our experience, that, with the rabbit, it is just the red muscles, which are distinguished for slowness in contraction, that contain less creatine than the white ones. Though the opinion of Bottazzi that muscular fibres show the phenomenon of tonus the more, as they are richer in sarcoplasma, as has already been pointed out by Mosso, is not quite in keeping with the observations, it may, however, especially after Engelmann's important researches, be assumed that the rapid contraction is performed by the anisotrope elements, accordingly by the muscular fibrils. The seat of the tonus must therefore be sought in the sarcoplasma or perhaps in the parts of the fibrils between which the anisotrope elements find a place. In a further investigation into the two different kinds of contraction of the muscular fibres it will certainly be of importance to keep the attention also directed to the double innervation again demonstrated by Boeke.

As to the starting-point of our investigation we think we are entitled to give an affirmative answer to the question whether the formation of creatine, and consequently the consumption of protein in the body, is largely influenced by the tonus of the muscles. Already many years ago it was proved by Pflüger [1]) of how great an importance the muscular tonus is for the production of heat. If our opinion is correct, it also follows from this that limitation in the supply of protein with the food, which is at the present day aimed

[1]) Pflüger's Archiv., Bd. XVIII, S. 247.

at by many, has its dangerous side. Mechanical labour the muscles can perform at the cost of food free from nitrogen; however to be of service to the organism, also in other respects, by means of the tonus, they want protein.

The opinion has often been pronounced that the stiffening of the muscles after death should be considered as a last contraction of the muscles. Especially HERMANN has indicated the agreement between the changes the muscle undergoes at coagulation and those which are observed in the contraction. In the above mentioned paper of VON FÜRTH and SCHWARZ it is proved that it is such substances especially, which are capable of promoting the coagulation of the muscle-plasma, that raise the labouring-faculty of the muscles.

It seems that the agreement does not refer to the rapid contraction but to the tonus. We have found an increase of the quantity of creatine in frog-muscles which were stiffened by immersion in water of 42° or 45° C. In four experiments the increase amounted on an average to 0.305 mgr. creatinine on 1 grm. of the muscle (min. 0.204, máx. 0.460 mgr.).

For the rabbit the investigation offered some difficulties, because here the decomposition of creatine, proved by GOTTLIEB and STANGASSINGER plays an important part and the so much thicker rabbit-muscle is not so rapidly coagulated as the thin muscles of the frog. When, however, the errors arising from this are avoided as much as possible, also in the majority of cases, both with the red and the white muscles of the rabbit, a distinct increase of the quantity of creatine was observed in the stiffened muscles.

Also in the investigation into the spontaneous stiffening of muscles after death, the postmortem disappearance of creatine has to be taken into consideration. When, however, the muscles of one side of the body were, directly after death, put in hydrochloric acid and the corresponding muscles of the other side after three or four hours when the stiffening had been well developed, each time there was found more creatine in the coagulated muscles than in those examined in a fresh condition. In the four cases dealt with in this way, we found an uncommonly great difference in one, and in the three others on an average 0.260 mgr. of creatinine more (min. 0.124, max. 0.336 mgr.). The description in details of these and the other observations mentioned we intend to give somewhere else.

From our investigation we think we are entitled to derive that in the muscles of vertebrate animals, at the heat-coagulation and the postmortem rigour as well as the tonus, a chemical process takes place which causes the origination of creatine.

Botany. — *"Sap-raising forces in living wood."* By E. REINDERS.

(Communicated by Prof. J. W. MOLL.).

Of the many theories, which have been advanced in explanation of the transpiration-current of trees, most are at present only of historical importance in the literature. The imbibition theory of SACHS [1]); BÖHM's atmospheric pressure theory [2]); the gas pressure theory of HARTIG [3]); the views of WESTERMAIER [4]), who regarded the xylem parenchyma as the water conduit and considered the vessels to be reservoirs; EWART's [5]) hypothesis that the living elements help te overcome the resistance, the cohesion theory of ASKENASY [6]), which neglected to adopt the continuity of water as a conditio sine qua non — all these have been given up. On the other hand opinion is still divided with regard to two hypotheses, the advocates of which combat the views of their respective opponents with remarkable asperity. GODLEWSKI [7]) and his supporters defend the view that the transpiration-current cannot be explained without postulating the cooperation of the living elements of the wood; DIXON and JOLY [8]) on the other hand advance the proposition that the living elements have not, and cannot have, anything to do with the process. They explain the phenomenon that water ascends up to the summits of the highest trees by assuming that in these trees the water, enclosed in the narrow water conduits, hangs like a thread from the surface of the leaf cells, where it is held by capillary or other physical forces. The thread does not break, because, as is supposed, it is nowhere in contact with air, and in these circumstances water can support a tension of 150 atmospheres. When the water evaporates in the leaves at the summit, this thread is drawn up through the tissues.

The keenness with which the two parties oppose each other is best illustrated by a couple of quotations.

SCHWENDENER [9]), an advocate of the more physiological theory, says:

"An der Vorstellung, dass die Lebenstätigkeit der Zellen irgendwie in die Saftbewegung eingreift ist . . . . unbedingt festzuhalten. Ohne dieses Eingreifen ist die Hebung des Wassers auf Höhen van 150-200 Fuss und darüber einfach unmöglich und alle Bemühungen, die vorhandenen Schranken mit unklaren physischen Annahmen zu durchbrechen, sind nicht viel mehr als ein Suchen nach dem Stein der Weisen".

In the same year 1909 DIXON [10]) writes:

"The adhesion of writers to the vital hypothesis . . . . is so

remarkable that we must devote some space to examine fully the grounds for their contention".

When we attempt to trace why opinions diverge so widely, the cause seems to lie principally in a different appreciation of certain experiments and in the somewhat adventurous aspect which the Dixonian explanation presents at first sight. It is necessary to become accustomed to the idea that the life of our trees hangs upon a water-thread, before we can become reconciled to it. GODLEWSKI [11]) indeed required a much more adventurous hypothesis in order to reconcile the anatomical structure of the wood with its power of pumping up water. This part of his theory has in consequence received adhesion from no one and so I will leave it out of discussion. In what follows below, "GODLEWSKI's theory" will therefore mean the view that the living wood must be regarded as the cause of the transpiration current.

In order to facilitate a judgment of the state of affairs I will tabulate the most important arguments of the two parties side by side and will then discuss them in pairs. From this table I omit everything relating to the question whether the cohesion of water is sufficiently great to account for the work which DIXON and JOLY attribute to it. I will assume, if I may put it thus, that there is no technical objection to their theory and I think this assumption may be made with safety.

GODLEWSKI c. s.	DIXON and JOLY.
1*a*. There is not sufficient continuity in the water columns of the wood to admit cohesion as an explanation [12]).	1*b*. There is no reason for doubting the continuity of the water columns [13]).
2*a*. The remaining available physical forces are insufficient to raise the water more than 14 metres [14]).	2*b*. STRASBURGER's experiments in which the water ascended in poisoned trees, prove the contrary [15]). The cohesion theory has at its disposal forces which would be able to provide a tree of 200 metres and more with water [16]).
3*a*. URSPRUNG's experiments, with branches which had been killed for part of their length, after which the	3*b*. In URSPRUNG's experiments the conduits become blocked and the leaves were poisoned because

leaves faded, prove that dead wood cannot transport enough water to balance the transpiration [17]).

4*a*. The structure of the wood is in favour of GODLEWSKI's theory [22]).

5*a*. Arguments from analogy [22]).

6*a*. The distribution of pressure in living transpiring trunks is opposed to the cohesion theory [23]).

they got a decoction of wood for their drink [18]).

4*b*. "The very structure of the wood offers the strongest evidence against GODLEWSKI's theory" [19]).

Living wood offers the same resistance in either direction to the forcing through of water [20]).

5*b*. Arguments from analogy [21]).

6*b*. The measurements of pressure are considered unreliable or are left out of account.

Point 1. The question of the "continuity of the water-threads" in the wood amounts to the following. The cohesion theory requires the assumption that the water in the tree forms one connected mass from the root to the leaves. Every xylem vessel in which there is an air-bubble has according to this theory become useless for the conduction of water, for in such a vessel the water cannot be under negative pressure; it is at once sucked empty by the adjoining vessels. Every bubble of air therefore puts one vessel out of action.

Now if it could be shown that by far the largest proportion of vessels contain air bubbles, only a small percentage would remain available for the conduction of water, and perhaps here and there the required connection of the water would be entirely interrupted, so that there could be no question of the cooperation of cohesion.

It is of course difficult to prove the absence of air, for in the necessary manipulations preparatory to the examination there is always the chance that air bubbles in some way or other get into the vessels [24]). If air is found in the majority of the vessels this does not prove that it was already present in the living plant, for it may have penetrated during manipulation.

For the further course of my argument it matters little, however, whether DIXON and JOLY or whether their opponents are right on this point. I will not therefore discuss it any further.

Point 2. The proposition, that physical forces alone are insufficient [25]) to raise water higher than 13—14 metres is a very weak point in the defence of GODLEWSKI's theory, for STRASBURGER's intoxi-

cation experiments have proved in the most striking manner, that this proposition is untenable. He found that water still ascended to the highest tops of the p o i s o n e d trees, up to a height of 22 metres.

The attempts of GODLEWSKI's supporters to maintain their proposition in spite of this fact give a very unsatisfactory impression. STRASBURGER is attacked in vague terms [26]); he is accused of a want of critical insight, he is reproached for not making any attempt at explanation: the fact itself remains.

The following argument appears to be somewhat more weighty. It is said [27]): "with the help of a JAMIN chain atmospheric pressure may be imagined to force water up to 13—14 metres"; but fourteen is not twenty-two and moreover a JAMIN chain can by no way explain anything in this case. It might perhaps be applied to this purpose with some chance of success, if the vessels ran through continuously from the root to the leaf, but certainly not in a system of vesicles like the wood, where the bubbles cannot pass the partitions, dividing up the conducting tracts, to say nothing of the multitude of other clinching objections.

It is further adduced against STRASBURGER, that continuous liquid threads are formed when the trunk, having been sawn off, is placed in water [27]), but in the first place it is not clear what objection is really meant by this and in the second place it is difficult to imagine how these threads are supposed to originate. The water which is sucked up cannot remove the air present, for the air is enclosed; it is moreover saturated with air, and is more likely to give off bubbles than to absorb them, as soon as it is exposed to a lower pressure at a certain height. Sawing off the tree will hardly affect its air-content except to increase it; the air which enters does not, however, endanger the cohesion, as it cannot ascend.

P o i n t 3. URSPRUNG's experiments [17]) with branches, which had been killed by steam over part of their length, in consequence of which the leaves faded, do not prove much for GODLEWSKI either. The steam not only kills the living elements, but also induces other changes.

For some time the vessels must conduct a decoction of wood instead of water and a blocking of the membranes or even of the lumina of the vessels may be the consequence, so that the resistance increases. The cells of the leaves are further more or less poisoned by this liquid, so that it is very doubtful whether the death of the leaves may be attributed to a want of water [18]).

These experiments are therefore not of much importance in deciding the question under consideration.

Point 4. The anatomical structure of the wood is a better argument for DIXON [19]) [20]) than for GODLEWSKI, for as yet it is quite impossible to imagine in what way the living elements could really exert any successful pumping action. The unidirectional resistance without which such an action can hardly be conceived, has never been observed, in spite of a careful search for it.

This argument is therefore no longer always adduced in support of GODLEWSKI.

Point 5. In critical cases the arguments from analogy are hardly more valuable than illustrations. I will therefore not discuss them here.

We see therefore that the arguments which have been advanced so far give little support to GODLEWSKI's theory. On the other hand the striking and conclusive result of STRASBURGER's intoxication experiments is in favour of DIXON and JOLY. If to this be added the great convincing power which proofs from analogy exert, when well presented (and here DIXON and JOLY are much more fortunate than their opponents), we may readily understand, that the cohesion theory has many supporters.

There are, however, two facts which are adduced against this theory with more success.

In the first place a second series of experiments by URSPRUNG [18]) in which he used ice instead of steam, in order to render part of a branch inactive. This series of experiments does not of course suffer from the objections which deprived the other series of its argumentative value. The fact, however, that fading only occurs after several days, makes the result less convincing.

Another objection is more important:

Point 6. The distribution of pressure in living trees is opposed to the theory [23]).

In a hanging water-thread the pressure decreases gradually as one ascends and the decrease is at least one atmosphere for an ascent of 10 metres. In living transpiring trees it has been impossible to demonstrate this; it was found on the contrary that manometers placed at different heights up the trunk, behave quite independently of one another. Sometimes one shows a lower pressure, sometimes the other.

It is true that objections can be raised against many of these measurements of pressure, but some of them in SCHWENDENER's opinion proved positively and undeniably that there can be no question of a regular decrease of pressure. For in this case it would be inconceivable, "dass ein Baumstamm der nach 2—3 Regentagen durch Nachschub von unten etwas wasserreicher geworden, in mittlerer

Höhe (wo vorher Saugen stattfand) Luft in das hier angebrachte Manometer hineinpreszt, während oben in der Krone und insbesondere unten am Stamm weder Saugung noch Pressung stattfindet" [23]).

It is remarkable that DIXON, in his review of the state of the problem in the "Progressus", does not at all refer to the ice experiments of URSPRUNG, nor to measurements of pressure, although he there considers at length and refutes much less important objections.

Thus we have traced the causes of the remarkable phenomenon mentioned in the introduction. The partisans of GODLEWSKI point to the measurements of pressure and maintain that STRASBURGER's experiments are invalid, whereas DIXON points to STRASBURGER and is not concerned with pressure measurements.

As will be seen the position is somewhat confused. In my opinion no advance can here be made along a theoretical road. Experiments alone can lead us out of the confusion.

I think I am able to supply conclusive, experimental proof that the normal living wood is able to pump water actively.

In order to give this proof I started from the following preliminary conception. If the irregularity of the results of pressure measurements is really caused by a pumping action of the living wood, this irregularity must at once disappear as soon as the experimental trees are killed or paralyzed. This was indeed found to be the case. Moreover, as soon as the trunk was dead the differences of pressure followed the same rule as would be expected to apply to a glass tube. When the conditions became unfavourable to evaporation, as in the evening and when rain supervened, the indications of the manometers approached each other more and more. At midday, in sunshine, on the other hand they differed more. This becomes intelligible, when we consider that a more rapid evaporation requires a stronger current; for a stronger current larger differences of pressure are however necessary, in order to overcome the greater resistances.

First I will describe the experiments somewhat more in detail. Later I hope to publish the curves of the positions of the manometers, together with the result of a more extensive investigation of this subject.

Of a $\pm 2\frac{1}{2}$ metres high specimen of *Sorbus latifalia*, which divided a little above the ground into two almost equal, strong branches, one branch was left intact as a control; to the other I fixed above one another some U-shaped open mercury manometers, in the following manner. Some lateral branches were cut off from the main branch in such a way that a stump of 5 cm. length remained. A tube was slid over this stump, and to it the manometer was afterwards fixed

this tube was blown out in the middle to a small bulb, and was hermetically fixed to the stump with a piece of india rubber tubing. It was then half filled with water, and momentarily pumped empty that we may inject the cut vessels. I then left it open for half an hour and finally closed it with the perforated rubber stopper, through which the manometer was stuck. Once a day the bulb-tubes had to be replenished, for the wood always leaks a little from the intercellular spaces. The bark leaks still more and for this reason I always removed it at the place where the rubber tube was to come.

As long as the tree was alive, no regularity could be perceived in the indications of the manometers: they all showed a pressure, smaller than that of the atmosphere, but sometimes one "sucked" more, sometimes another. After a few days I killed the portion of the branch bearing the manometers over its whole length by means of steam. At once the manometers followed the rule indicated above, and did not depart from it. The differences of pressure became very considerable towards midday, showing that the dead portion offered a great resistance to the strong current.

The crown and the base of the branch remained intact during this treatment. The leaves showed only after 3 weeks, that they had suffered from the operation; up to that time they remained perfectly fresh. When at last they began to change, they gave the impression that they were diseased, rather than that they suffered from want of water.

Two manometers were attached to the small trunk of a *Cornus* and fixed to almost equal stumps of branches, the one 66 cm. above the other. The whole tree was 2 metres high. Before I cut off the branches, which were to yield the stumps, I killed the trunk at these two places with steam over a length of 10—12 cm. The manometers were thus attached to dead branch stumps on dead pieces of the trunk, separated by a living portion.

I wished to investigate whether the living intermediate portion did pump or not. If so, it would always be occupied in diminishing the difference of pressure between the two dead pieces of the trunk. If it was then suddenly cooled with ice, the manometers would have to diverge suddenly and would once more approach each other if the tree was left to itself. Finally if it was killed, the well-known regularity would be bound to appear.

The result was different, however. The intermediate portion evidently did not pump, for the manometers behaved exactly as in a dead tree. At midday they sometimes differed by 24 cm. of mercury. However — on the fifth day their behaviour changed fairly

suddenly and on the sixth day it was as irregular as in living trees!

Evidently the intermediate portion had suffered too much by this treatment, to function immediately, but on the sixth day it had so far recovered, that it could work again. It lived on until the end of December, as could be seen by the perfectly fresh bark. Now, at the end of January, it is dead. The crown, however, still looks healthy, as also do the buds.

Although those facts, as far as I can see, do not permit of an explanation other than the one given here, a proof may still be adduced that such phenomena cannot be attributed to a change in the resistances. Such a change would moreover have to be of a very remarkable nature to be of any use as an explanation.

Four manometers were attached to the trunk of a lilac tree (*Syringa vulgaris*) 2 metres in height, and they were numbered in ascending order 1, 2, 3, and 4. After a short time they all showed an approximately equal "suction", which oscillated with diurnal periods between 48 and 28 cm. of mercury. Although the differences were small, some times one was the highest, some times another. After 15 days, when I knew the course of the pressure curves sufficiently, stump 2 was killed, together with the piece of the stem to which it was attached. This was done by passing through it for an hour the discharges of an induction coil capable of giving a spark of 10 cm. long, without sparking in the secondary circuit. The stump and the portion of the trunk became heated to nearly 60° C.: a few pieces of glass cement of that melting point, which I had fastened to it, just began to melt.

While the induction current was being passed, the suction of stump 2 first diminished greatly, as a result of the heating, the other manometers remained constant. Soon the fall of the mercury in no 2 stopped and the suction increased again. After the interruption of the current the mercury rose higher than usual; this abnormally high suction subsequently persisted; no 2 afterwards followed the periods of the other manometers, which went on without hindrance, but sucked always strikingly more. How we can deduce from this the proof that this phenomenon is not caused by changes in the resistances, will be explained presently.

Thus far the description of the experiments. I will now consider what may be deduced from the results.

The course of the manometers in *Sorbus* proves that the water current in a living tree is caused by quite different forces from those of a dead one. The result cannot be attributed to the imperfectness of measurements. Most of these are the same before and after death

and we cannot suppose that the circumstances which are changed in the operation, are altered exactly in such a way as to bring to light the observed regularity. Thus the distribution of pressure before death can only be explained on the assumption that there are pressor factors, i.e. pumping actions in the wood.

This view receives important support from quite a different side, through the experiments of ZIJLSTRA [30]). He allowed a solution of Säureviolett to ascend living and dead branches and then examined them microscopically. In the living ones only the tori of the bordered pits were stained, together with a thin layer of the walls of the vessels; in the dead ones, however, the whole of the wood was coloured uniformly. It follows from this that the water current takes quite a different course in dead wood from that taken in living wood.

That in the lilac only the one manometer was affected, which was attached to the portion killed by induction shocks, cannot in my opinion, be explained in any other way than by the aid of GODLEWSKI's theory. If one imagines, with DIXON and JOLY, that the whole trunk behaves like a dead tube, the phenomenon cannot be explained. An increase of resistance cannot be the cause, for then the other manometers would have undergone this influence. If on the other hand, we imagine a tree-trunk to be a system of tubes in which everywhere small pumps occur, the phenomenon becomes intelligible. The death of the piece of trunk puts the pumps out of action locally and the suction must there be somewhat greater to get the water through the piece of dead wood. This would not necessarily be observed at the following manometers, since the intermediate elements bring the pressure back to normal.

I regard all the above as proof positive that the living wood has a hydromotory power. The experiment with *Cornus* already proves this very clearly: one could almost see the recovering intermediate portion suddenly begin pumping, as it were before one's eyes.

After thus having given the positive proof that the living wood assists in the ascent of the water I will again take up the theoretical considerations with which I started, and see to what extent this proof can modify the condition of affairs.

We encounter the difficulty that STRASBURGER's intoxication experiments prove that help of living elements is not necessary, whereas the only theory which is not adversely affected by these experiments becomes untenable on account of the pressure measurements. The solution is clear from the preceding.

The adherents of GODLEWSKI are wrong in asserting that water

cannot ascend more than 14 metres without the help of life, for STRASBURGER's experiments show that this is indeed possible. But that is not the question. The position is simply this, that in a living tree the water is pumped up by living elements, whereas in a dead one it also ascends, but through other causes e.g., with the help of cohesion.

Let us test this view by the data in the table:

Point 1. The question of continuity is only of importance for the cohesion theory. As soon as this has been refuted in another way, the question of the continuity of water, may be left untill it may perhaps arise again in connection with new questions.

Point 2. The intoxication experiments of STRASBURGER have been included in my thesis.

Point 3. Although the experiments of URSPRUNG do not prove anything certain in favour of GODLEWSKI's theory, they certainly prove nothing against it.

Point 4. The anatomical structure of the wood can never be adduced as an actual objection to the view here put forward. As soon as it has been proved that the living wood pumps, this fact cannot of course be weakened because we cannot at once imagine from its structure how this action may take place. The investigation of this point must simply be left for further research.

Point 6. The distribution of pressure is quite in agreement with GODLEWSKI's view. When pressor factors are everywhere present in the trunk, the distribution of pressure cannot be predicted as long as these factors themselves are not fully known.

We see therefore that the questions discussed here do not produce an objection to my view. In this preliminary communication I have of course limited myself to the most important matter; afterwards I hope to treat the same subject more completely.

I might have omitted the literature entirely, but it seemed desirable briefly to justify my quotations and references. The small figures in the text refer to the bibliography which is appended below.

I wish to conclude this preliminary communication by pointing out that the method which is here introduced, may also be of service in the solution of other questions. By its aid we might, for instance, ascertain whether the living elements cooperate, when a branch is made to transport water in the inverse direction; the influence of all sorts of stimuli (heat, cold, electricity, stimulant substances) on the activity of these elements can be examined. Should the intoxication experiments of STRASBURGER be repeated with manometers fixed to the experimental trees, they would at once constitute a

definite proof in favour of GODLEWSKI. A small tree would, however, be sufficient for this.

The method in which a living piece of wood is isolated between two dead portions is especially to be recommended. The portions to be killed should not, however, be heated above about 60°, in order to spare the intermediate position (compare the experiment with the induction current). If possible a leafy branch should be left attached to the intermediate portion, for otherwise it must soon die of hunger.

Groningen, January 28th 1910.

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Botany. — "*Contributions to the knowledge of the movement of water in plants.*" By Dr. K. ZIJLSTRA. (Communicated by Prof. J. W. MOLL).

(Communicated in the meeting of January 29, 1910).

For some time I have been occupied in the botanical laboratory at Groningen with the problem of the movement of water in plants and have carried out experiments of a somewhat diverse nature. Various circumstances have prevented me from continuing my experiments in this direction, so that the investigation has not been rounded off. I did not intend publishing it, but as I shall presumably have for some time no further opportunity of continuing my studies, I think I may be justified in publishing the data I have collected; possibly they may be of service to other investigators who have chosen for their researches the subject of the movement of water in plants.

The experiments referred to may be arranged under three heads, viz.:

1st. The trunk or stem of intact plants cooled to about 0° C.

2nd. The ascent of a dye solution in cut branches.

3rd. Interference with the movement of water in a tree-trunk by means of deep incisions.

First I propose to discuss the considerations which led to these experiments and the results obtained, and then I will give a more detailed account of the execution of the experiments.

1. *Trunk or stem of intact plants cooled to about* 0° *C.*

As is well known, GODLEWSKI (Zur Theorie der Wasserbewegung in den Pflanzen. Jahrb. f. Wiss. Bot. Bd. 15) attempts to find the cause of the movement of water in the activity of the living cells of the medullary rays and of the wood parenchyma; these cells would therefore have to act as it were as suction-pressure pumps. GODLEWSKI did not, however, adduce any direct experimental evidence in support of this theory. His theory is only made plausible with the aid of various data obtained by others, and it is urged that the theory does not conflict with the facts adduced by other investigators. Various botanists (JANSE, STRASBURGER, WEBER, URSPRUNG) have afterwards attempted to test the theory experimentally.

The most obvious method for such a test would be the following: to cut out the action of the living cells of medullary rays and wood parenchyma, and then to see whether the movement of water had become impossible.

This elimination of the action of living cells was most easily obtained by simply killing these cells by poisons or by a high temperature.

This method is, however, open to objection; such interference not only attains the elements which it is desired to put out of action; others also, especially the water-conducting vessels and tracheids will undoubtedly be affected, so that it is questionable whether the results of the experiments can only be attributed to the elimination of the activity of the living cells.

A method — already used by URSPRUNG but with a result opposite to mine — which meets this objection, is the cooling of the trunk or stem of the plant to about 0°. By this means it is possible to reduce the activity of the living cells to a minimum, while neither dead nor living elements undergo a permanent change. Moreover the advantage of being able to establish the original conditions after the conclusion of the experiment and therefore bring the plant back to normal conditions, should not be underrated. The experiment and its control can both be carried out on the same intact plant.

If by this means the plant could be made to fade, and to assume its original fresh appearance after the cooling had been stopped, GODLEWSKI's theory would receive considerable support.

According to this method I have myself carried out 3 experiments. The trunk of a small apple-tree, 2 stems of *Polygonum cuspidatum* and 2 stems of *Helianthus tuberosus* were cooled to about 0° C. over a length of 50 cm. The experiments lasted 6, 7, and 8 days, under conditions which were very favourable for a possible fading.

Nevertheless I have in no case been able to observe even incipient fading, although the transpiration from the leaves was strong, as shown by the cobalt test. Cut leafy branches, hung up near the plant, withered very rapidly.

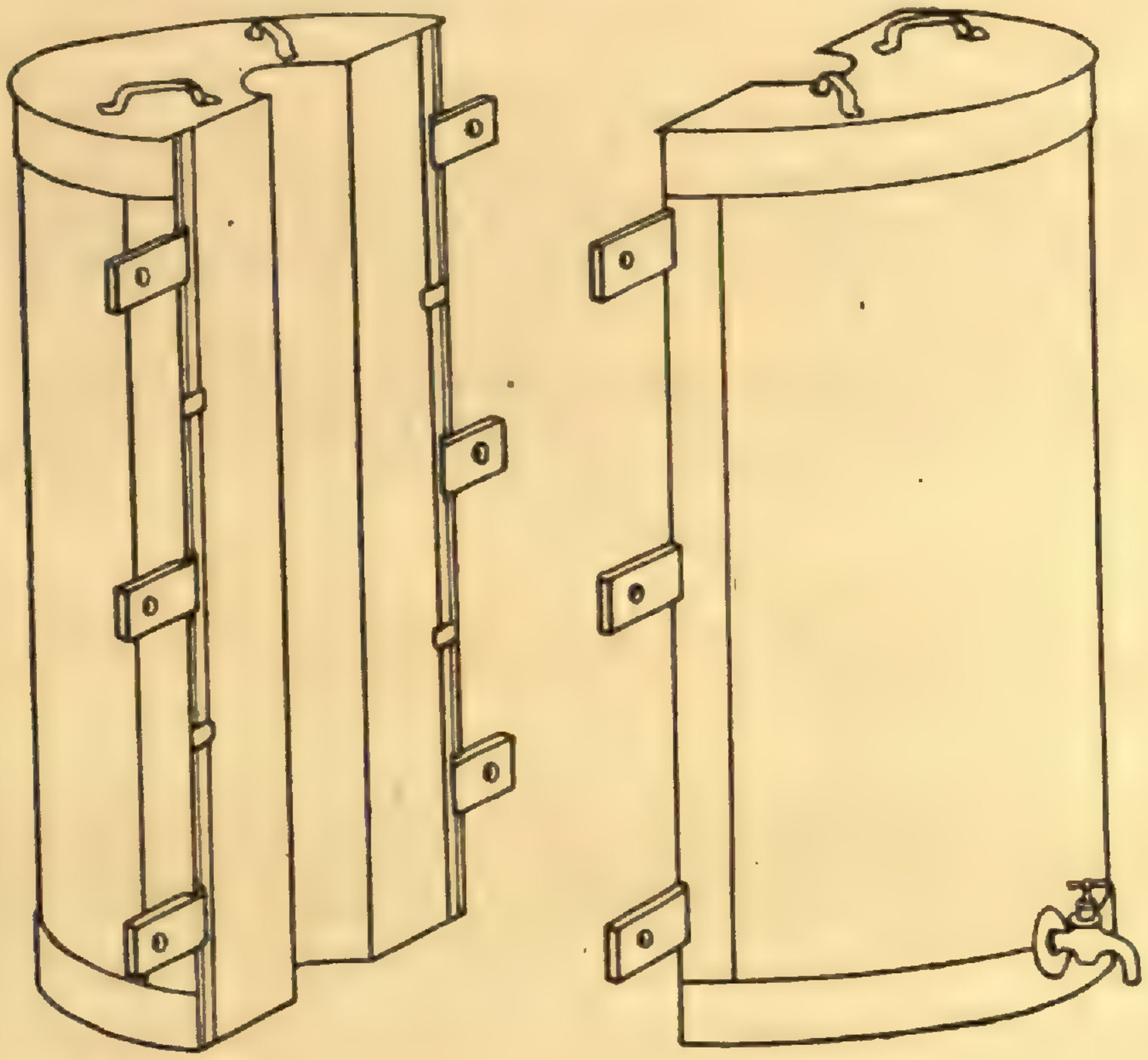
We may not, however, conclude from the negative result of these experiments that the living cells do not play a part in the movement of water. It is quite possible, even probable, that cooling a length of 50 cm. is not enough. This slight obstacle was perhaps easily overcome by differences of pressure present in the trunks. Had the results been positive it would have supported GODLEWSKI's theory. My negative results are, however, not able to oppose this theory.

The nature of the results notwithstanding, I think it may be useful to bring them to the notice of others.

Description of the experiments.

The cooling of the trunk or stem was brought about by melting ice, which was placed in an apparatus indicated by the figure.

The apparatus consisted of two equal parts, i.e. of two semicircular tin-plate reservoirs with fixed bottom and loose lid. The two half-



cylinders had on the middle of the flat side a portion which was bent like a half cylinder, so that the two reservoirs when joined to form one cylinder, left in the centre a space for the passage of the trunk, which was to be cooled. The height of the apparatus was 50 cm., its diameter 30 cm.; the space left free for the trunk had a diameter of 10 cm. Each reservoir was provided at the bottom with a tap, through which superfluous water could run off. The cylindrical surface of each reservoir and also the bottom and the lid, were covered on the outside with a layer of felt, 15 mm. thick and over this there was a covering of asbestos paper, 2 mm. thick. In an experiment the two reservoirs were placed round the stem and screwed together, after a piece of felt had been placed between the two flat surfaces in apposition.

The reservoirs were filled with ice. The space through which the trunk passed, was closed off above and below round the stem by a solid plug of cotton wool through which a thermometer passed. The temperature in the annular air space surrounding the trunk varied between 0° and + 3° C.

The apparatus was sufficiently protected by the felt and the asbestos against the heat of the surrounding atmosphere. Even on hot days it was only necessary to renew the ice twice in 24 hours.

During the experiment the apparatus rested on some bricks, so that it was about 20 cm. from the ground.

Experiment I.

Apple tree.

The ice apparatus was fixed round the trunk having a diameter of $3^1/_2$ cm., of a small apple tree, about $2^1/_4$ metres high, on July 21st 1904, at noon, the weather being hot and sunny. The apparatus was filled with ice and in the course of the afternoon the temperature in the space round the trunk fell to about 1° C.; not the slightest fading of the leaves could be detected, although such fading would have at once been noticeable by comparison with two other apple trees which stood next to the tree experimented on.

Nor could any change be observed on the following days. The temperature of the air space round the stem remained continuously between 0° and 3°.

The maximum temperature of the atmosphere on the days of the experiment oscillated between 23° and 29°.

On the sixth day, when the temperature of the atmosphere was 20° and that round the trunk 0°, an strong transpiration of the leaves was demonstrated by means of the cobalt test. On the seventh day the trunk was sawn through immediately above the ice apparatus; a hole was drilled in the portion of the trunk still inside the apparatus, and a thermometer was placed in it.

In this way I was able to show that the temperature *inside the trunk* was the same as that of the annular air space *round the trunk*, i.e. in the course of three hours it oscillated between 2° and 3°, while the temperature of the atmosphere was 24° to 25°.

Experiment II.

Polygonum cuspidatum.

The ice apparatus was fixed round two immediately adjoining stems, 2 metres in height, on July 6th 1905, at noon, and it was filled with ice. In the course of the afternoon the temperature in the air space round the stems fell to 0°, without withering taking place. The numerous stems surrounding the apparatus served as controls. Nor was any change noticeable during the following days.

The temperature round the stems remained continuously between 0° and 3°. The maximum temperature of the surrounding atmosphere

on the various days during the experiment oscillated between 19° and 30°. The experiment was stopped on the seventh day.

Experiment III.

Helianthus tuberosus.

The ice apparatus was fixed round two immediately adjoining stems of plants, $1^1/_4$ M. in height, on July 14th 1905 at noon, and was filled with ice. In the course of the afternoon the temperature of the air space round the stems fell to 0°. No fading could be observed; several specimens of the same species, standing next to the plants experimented upon, served for comparison. Cut leaves, hung up on the plants were completely withered in a few hours. Nor was withering observable on the experimental plants on following days.

The temperature in the air space round the stems remained about 0°. The maximum temperature of the atmosphere in the days of the experiment oscillated between 17° and $26^1/_2$°. The experiment was stopped on the eighth day.

2. *Ascent of a dye solution in living and dead cut branches.*

When cut branches, with a freshly cut surface, are placed with this surface in a dye solution, the liquid will in general ascend into the branches for some distance, and thus may be easily traced by cutting them across at different levels. Various elements of the wood are then found to have been stained. It matters little whether one takes for this experiment living or dead branches, with or without leaves; the fluid always ascends in the branches, even when these are upside down, i. e. are placed in the solution with their cut apex. I generally carried out such experiments with twigs 30—40 cm. long; sometimes with pieces of a branch, which had also been cut at its upper end. After some days the stain shewed itself on the surface of the upper section of these latter branches.

Although the dye ascends in all branches, the *way* in which the various elements are stained is not the same in living and dead branches. A sharp difference is observable.

In comparing living branches with dead ones, it was of course necessary to use a harmless stain; the experiments of Stracke: investigation of the immunity of the higher plants towards their own poison (Dissertation), led me to choose *Säureviolett* of Grübler. I used this stain in a $^1/_{10}$ % aqueous solution. The twigs were placed

separately in a small bottle with the solution of the stain, the neck of the bottle being closed with a plug of cotton wool to prevent evaporation.

After the experiment the twigs were examined at different levels by microscopic sections. Transverse, radial and tangential sections were examined in *oil of cloves*, a medium in which Säureviolett is insoluble, so that the stain remained properly localized. The sections were cut without the use of any liquid and were at once placed in the oil of cloves. The slight water content of these preparations did not interfere. After a very short time the oil had thoroughly permeated. This method had moreover the advantage, that after most of the clove-oil had been wiped away, the preparations could be very well enclosed in Canada balsam, without further treatment.

A comparison of the behaviour of the xylem elements of living and dead branches brought out the following differences:

living branch	*dead branch*
a. torus of the closing membrane of the bordered pits deeply stained.	*a.* torus not stained, or only very slightly.
b. adjoining the lumen, a thin layer of the wall in the border of the pits is stained. The walls of vessels and fibres are only stained in a very thin layer, which is immediately adjacent to the lumen.	*b.* the walls of the vessels, fibres and parenchymatous cells are stained uniformly.
c. contents and wall of the cells of medullary rays and wood parenchyma are unstained.	*c.* contents of the cells are coloured.

The deep staining of the tori in living branches was especially noticeable, also in transverse sections, the more so because the staining of the layer next to the lumen in the walls of vessels and fibres was often difficult to see and because the living cells of the medullary rays and parenchyma were quite colourless.

In the wood of *Salix* and of *Fagus*, in which the tori cannot otherwise be seen at all, they were made very obvious by this staining of living branches.

The staining of the tori by eosine in a living branch of *Ginkyo* was already mentioned by JANSE in "Die Mitwirkung der Markstrahlen bei der Wasserbewegung im Holze" (Jahrb. f. Wiss. Bot. 1887 Bd. XVIII). In this case also the stain had ascended the branch:

the "primäre Wandlamelle" of the medullary ray cells was according to JANSE all that had been stained.

In my experiments with living branches the staining extended not only to the tori of the vessels and fibres, but also to those of the *half* bordered pits between the medullary ray cells on the one side and the vessels and fibres on the other side; the *contents* of the medullary ray cells however remained colourless, as stated above.

The results of other experiments carried out by me, agree well with these facts. Instead of taking dead branches, I caused to ascend in living branches a $^1/_{10}$ $^0/_0$ solution of Säureviolett in strong alcohol, and also a $^1/_{10}$ $^0/_0$ solution in water containing 4 $^0/_0$ formaldehyde. As controls I employed living branches in a $^1/_{10}$ $^0/_0$ solution of Säureviolett in water.

I now found that the living branches in the poisonous solutions were stained practically in the same way as the dead branches in innocuous ones, only not so completely. It was clear that the alcohol and the formaldehyde only gradually exercised their fatal action on the plant. The tori were always unstained; only a few were stained very faintly. The walls generally showed a uniform staining; the medullary ray and parenchyma cells with contents were coloured dark blue.

Finally I may add that microscopical transverse sections through living branches, which sections were afterwards placed for 20 hours in an aqueous Säureviolett solution of $^1/_{10}$ $^0/_0$, were stained quite uniformly dark blue, exactly in the same way as those sections made after the stain had ascended in *dead* branches; the colour was only somewhat more intense. The transverse sections through control branches, which had previously stood in the same solution for 4 days, on the other hand showed, as was to be expected, a staining quite similar to that which was described above for living branches.

Description of the experiments.

EXPERIMENT IV.

Fagus silvatica.

A living leafy twig, about 4 mm. thick at its base, stood for 9 days in a solution of Säureviolett. The stain ascended to the top and into the leaves. The bark, the cambium and the pith remained quite unstained; the staining was limited to the wood and here the stain was only in the inner layer (adjoining the lumen) of the walls of vessels and fibres; the tori of the bordered pits were stained a very

deep blue-violet, and this was also the case with the half bordered pits between medullary ray cells and fibres. The medullary rays and the xylem parenchyma were quite unstained, both as regards wall and contents.

Experiment V.

Larix decidua.

A living leafy twig, 6 mm. thick at its base, stood for 5 days in the solution, after which the stain had penetrated to the apex. Staining completely limited to the wood, but no stain in the oldest of the 6 annual rings.

The stain only taken up by a very thin layer of the wall, adjoining the lumen of the tracheids and the cavities of the pits. Torus of the pits deep blue-violet, also in the half bordered pits between medullary rays and tracheids. For the rest everything unstained.

Experiment VI.

Salix spec.

Two living leafless branches, provided at either end with a cut surface, both 30 cm. long and more than $^1/_2$ cm. thick, stood for 2 days in the aqueous solution of Säureviolett; one of the branches had its lower end in the solution, the other its upper end.

The stain ascended readily, and *in the two branches simultaneously*. The stain only present in a thin layer of the wall adjoining the lumen of the vessels and fibres and the cavities of the pits. Tori deep blue-violet.

Experiment VII.

Fagus silvatica. Taxus baccata.

Of each of these plants two similar 3—5 year old leafless branches were placed with the cut surface in the aqueous Säureviolett solution for 3 days. One of the branches of each species was alive, the other had been treated as follows. It had stood for $1^1/_2$ hours in boiling water. Then water was sucked through the boiled branch by means of a filter pump in order to remove possible obstructions, finally a fresh surface was cut.

After 3 days the stain had almost reached the top in all the four branches.

In the living branches staining was scarcely visible against the walls of the vessels and tracheids. The tori, including those of the half bordered pits were deeply stained. Medullary ray- und parenchyma cells quite colourless.

In the boiled branch of *Fagus* the walls of the libriform fibres and of the vessels were a uniform pale blue. Against the walls of the vessels in the spring wood a darker layer. Nowhere however coloured tori. The medullary rays also proved to be colourless.

In the boiled branch of *Taxus* the walls of many tracheids were stained a uniform pale blue; towards the inside against the walls a darker layer. The tori unstained. The medullary rays dark blue.

Experiment VIII.

Taxus baccata.

Two living branches were taken. One was placed with its cut surface in a solution of 0.1 gram of Säureviolett in 100 c.c. of *water*; the other in a solution of 0.1 gram of Säureviolett in 100 c.c. of *alcohol*.

Both branches remained standing in the solution for 43 hours, after which time sections were made through both at a height of 7 cm. The staining was as follows:

Branch in *aqueous solution*: staining only in the secondary xylem. A very thin blue layer against the walls of the tracheids, and of the cavities of the bordered pits. Tori dark blue, including those of the half bordered pits. Medullary rays unstained.

Branch in *alcoholic solution*: the stain had also penetrated into the cambium and the innermost layers of the cortex parenchyma, where both walls and contents were dark blue. In the secondary xylem the tracheid walls light blue; against the walls also clearly a blue layer, further in the cavities of the pits. Tori unstained. Medullary rays dark blue, both as regards walls and contents. The walls also coloured in the primary xylem.

Experiment IX.

Taxus baccata.

A living branch was placed with the cut surface in a solution of 0.1 gram of Säureviolett in 100 c.c. of a 4% *formaldehyde solution* (diluted formalin).

After 3 days the branch was examined; the stain had already reached the apex.

Staining only in the secondary xylem. Against the walls of the tracheids there was a thin blue layer, also in the cavities of the bordered pits. Tori colourless. Of the medullary rays both the walls and the protoplasm dark blue.

Experiment X.

Salix spec.

A living twig was placed with its cut surface in a solution of 0.1 gram of Säureviolett in 100 c.c. of a 4% *formaldehyde solution*. After 3 days the stain had penetrated to the apex and the twig was examined.

Staining only in the secondary xylem. The walls of the vessels coloured light blue with an indication of a somewhat darker layer adjoining the lumen. Tori practically colourless. The medullary ray cells, which adjoined the vessels, are coloured blue.

3. *Interference with the movement of water in a tree-trunk by means of deep incisions.*

Experiment XI.

An experiment with a small willow tree in the Botanic Gardens at Groningen showed, that in a trunk in which the transpiration current had been largely prevented or perhaps completely cut off as a result of transverse incisions on both sides at various heights, measures were taken in course of time which ultimately led to a complete recovery of this current.

The experiment was carried out as follows.

The trunk of the tree, $11^1/_2$ cm. thick, was sawn into transversely to slightly beyond the centre at four places, alternately on either side of the trunk. The incisions were 22 cm. apart, and the lowest was 1.25 Metres from the ground. They were prevented from closing up again by the insertion of tin plates, which in future remained in position. At these four places the water current was therefore irreparably interrupted.

As the trunk had of course been greatly weakened by this operation the tree was supported by four iron wires, which were attached high up to the trunk and also to four pegs driven into the ground at some distance round the tree.

This expeeiment was started on July 14th 1908; the incisions were ready and the plates were pushed in at 9.30 a.m. At 10 a.m. the leaves were already drooping and they remained so throughout the day.

In the course of the five following days, in cool dry weather, the leaves gradually recovered. On the 7th day of the experiment the foliage began to wither from the top downwards; many yellow leaves also appeared in the crown. In all these days the temperature had not risen above 18° in the neighbourhood of the tree. On the 9th day the temperature rose in the afternoon to more than 26°, and probably as a result of this the number of yellow leaves now increased rapidly. Those leaves which had remained green also began to droop again. The tops of the branches in the upper part of the crown withered completely.

The 3 following days were warm and sunny with temperature maxima of 27° and 28°. Most of the leaves now fell off, while in the upper half of the crown the foliage withered completely.

After this time cooler weather supervened and the few remaining green leaves recovered and remained in good condition until the autumn.

That the tree had not suffered greatly however from the incisions, was shown in the following summer, for then the foliage developed as well as before the experiment, and remained fresh throughout the entire season.

Wageningen, Dec. 13th 1909.

Physics. — "*The magnetic separation of absorption lines in connexion with Sun-spot spectra.*" (I). By Prof. P. ZEEMAN and Dr. B. WINAWER.

1. As a consequence of the intimate connexion between emission and absorption, there exists closely corresponding to the magnetic separation of emission lines, a magnetic division of absorption lines. The dark lines which appear in a continuous spectrum, if a beam of white light traverses an absorbing flame, are divided and polarized under the influence of magnetic forces in exactly the same way as the emission lines. This correspondence between emission and absorption was shown to exist already in some of the first experiments on the subject by one of the present authors. Our knowledge of emission spectra under magnetic influence has since been extended considerably. The experimental study however of the inverse effect i. e. the magnetic division of absorption lines has less advanced.

After the first experiments of the first named of the authors of this paper, the change of absorption lines in a magnetic field was studied by KÖNIG [1]) and COTTON [2]); RIGHI [3]) gave an elaborate study of the subject, to which we have to return later on. It contains the only investigation of the magnetic effect in a direction inclined to the lines of force. Closely connected with our subject are finally some observations by LODGE and DAVIES [4]) on the influence of a magnetic field on flames, emitting "reversed" lines.

The consideration of the inverse effect forms the basis of VOIGT's magneto-optical theories [5]); and it is considered also by LORENTZ [6]) in his investigation of the magnetic separation in a direction inclined to the line of force.

Theory indicates different points, which may be tested by experiment. The inverse effect has become of supreme interest in solar physics, since HALE's [7]) discovery that the dark lines of the sun-spot spectrum exhibit the characteristic phenomena of magnetic separation.

The experiments we intend to describe in the present communication relate to the division of the sodium lines D_1 and D_2. Some of our results may already be found in the work of the cited authors.

In order to present the subject in a connected form it seemed necessary not to exclude these.

The facts now ascertained in combination with former results appear to be of some value in explaining peculiarities observed in sun-spot spectra. Some instances will be given later on.

2. Type and relative amount of the magnetic division of the sodium emission lines, D_1 and D_2, are given in Fig. 1.

Fig. 1.

a represents the observations when the line of sight is at right angles to the magnetic field, *b* when it is parallel to the field.

In a weak magnetic field D_2 exhibits the triplet type, at right angles to the

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[1]) KÖNIG. Ann. d. Phys. Bd. **62**. 240. 1897.

[2]) COTTON. Éclairage Electrique. 5 et 26 mars. 1898.

[3]) RIGHI. Sul fenomeno di ZEEMAN nel caso generale d'un raggio luminosa comunque inclinato sulla direzione della forza magnetica. Mem. di. Bologna. 17 Dicembre 1899.

[4]) LODGE and DAVIES. Proc. R. Soc. **61** 413. 1897.

[5]) W. VOIGT. Magneto- und Elektrooptik. Chapter IV and the papers there cited.

[6]) H. A. LORENTZ. These Proceedings, Vol. XII, p. 321, 1909.

[7]) G. E. HALE. On the probable existence of a magnetic field in sun-spots. Contributions from the Mount Wilson Solar Observatory Nr. 30. 1908.

field ; the doublet type if the light is examined parallel to the lines of force. D_1 seems to exhibit a doublet in both principal directions.

The FRAUNHOFER lines in the spectra of sun-spots investigated by HALE are either broadened, or changed to doublets (often incompletely resolved quartets), or to triplets. The resolutions exhibited by sodium vapour are therefore the very types of special importance to astrophysics ; this and also the facility of producing sodium vapour in the magnetic field induced us to commence our experiments with this substance.

3. The explanation of the inverse effect is easily understood by means of the well known law of resonance. If there are in a flame under the influence of a magnetic field three periods of free vibrations, then we may expect that from incident white light vibrations of these very three periods will be taken away. The absorption is a selective one, with this peculiarity that the selection refers not only to the period but also to the direction of vibration. Consider for example the central component of a triplet which in the emission spectrum is due to vibrations parallel to the field. From incident white light only vibrations, corresponding as to period as well as to direction of vibration with the middle component, are absorbed. Vibrations, perpendicular to the field, though of the period of the unmodified line, pass unimpeded.

On the contrary white light of periods coinciding with those of the outer components is only deprived of its vertical constituents.

It will be clear from these very simple considerations what we may expect to observe with white light under the conditions of the experiment. The arrangement was the following: White light of the incandescent positive pole of an arc-lamp traverses a sodium flame, placed between the poles of a DU BOIS-electromagnet. This light is analysed by means of a stigmatic spectroscope with large ROWLAND grating. The observations are made in the first order.

If the observation is made at right angles to the lines of force, we see in the continuous spectrum 4 dark components in the case of D_1, 6 dark components in the case of D_2, as represented for both lines under *a* in the diagrammatical Figure 1.

In order to observe all these components the field must be strong and the vapour density adapted to the field.

The groups of lines indicated by *b* are seen, if the light is examined axially.

All these components, if narrow, are seen only diffuse and not black. From the considerations above given the reason will be clear at once; each of the components absorbs only *half* the incident natural light.

With very diluted vapour no absorption at all or only very weak traces of absorption are seen.

4. The introduction of a Nicol in the beam before or after the field entirely changes the phenomenon. The absorption lines can then be seen very narrow and black.

Let the observation be made at right angles to the horizontal field, then, if the Nicol is placed with its plane of vibration vertical D_1 exhibits its two, D_2 its four outer components.

After a rotation of the Nicol over 90° both D_1 and D_2 give only the two horizontally vibrating components.

Let a beam of natural white light traverse axially the magnetized vapour placed between the perforated poles of an electromagnet. Then by means of a quarter-wave plate and a Nicol we may quench either the right-handed or the left-handed circularly polarized component.

A combination of a quarter-wave plate and a Nicol, converting incident light into right-handed circularly polarized light may be called a right-handed circular analyser. The absorption line corresponding to a right-handed circularly polarized component is seen with both increased clearness and darkness by examining it with a right-handed circular analyser.

We introduce here this simple matter because there has been occasionally some confusion on this subject.

5. The behaviour of horizontal and vertical vibrations may be studied simultaneously by using according to the suggestion of Cornu and König a calcspar rhomb. By means of it we can obtain two oppositely polarized images of a horizontal slit of suitable width, placed near the magnetic field.

Right-handed and left-handed circular vibrations can be separated on the same plan by the introduction of a Fresnel rhomb between the calcspar and the slit of the spectroscope.

It is, however, of considerable interest to examine also the behaviour of the lines in natural light. A separate examination after the removal of the polarizers might be made. The vapour density ought to be the same in both experiments. It seems difficult to realise this.

The desired end is secured more simply and surely, and with only half the labour, by adopting the width of the horizontal slit and the thickness of the calcspar in such a manner that the two images given by the calcspar partially overlap. We now obtain three stripes; the central one exhibits the phenomena as seen without polarizing apparatus. (See fig. 2).

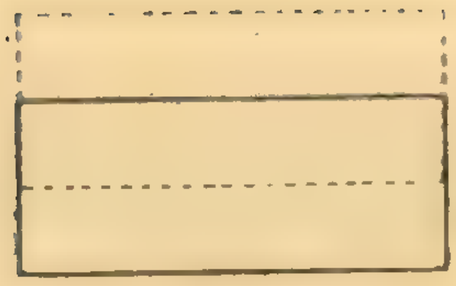

Fig. 2.

The upper and lowest stripes show the influence of polarized light on the phenomenon.

The observations given in this communication have been made by the described method. By its use all particulars of the phenomenon are simultaneously exhibited; we also succeeded in photographing the essential points. Examples of our photographs are given on the plates annexed to our paper.

6. If the absorption lines are not narrow or if the magnetic field is weak, the components of a magnetically divided line will partially overlap. This partial superposition is the cause of some particularities, especially manifest in the inverse effect and probably also apparent in sun-spot spectra.

The nature of these particularities may be illustrated by a few examples. We will consider the case of the magnetic triplet and the magnetic doublet.

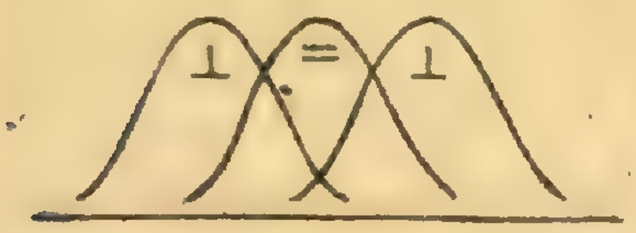

Fig. 3.

In Fig. 3 the curves show the distribution of intensity of the three components of a triplet, if the light is examined at right angles to the lines of force. If natural light traverses a source of light placed in a magnetic field, two black bands are seen, corresponding to the wavelength, for which vertical as well as horizontal vibrations are absorbed.

These black bands are surrounded by less dark parts, which absorb only one of the principal vibrations, the other proceeding unimpeded. (cf. §§ 3 and 4).

If now a Nicol with its plane of vibration vertical, is introduced two black bands are again seen. The darkest part of these components corresponds to the maximum of the curves relating to vertical vibrations.

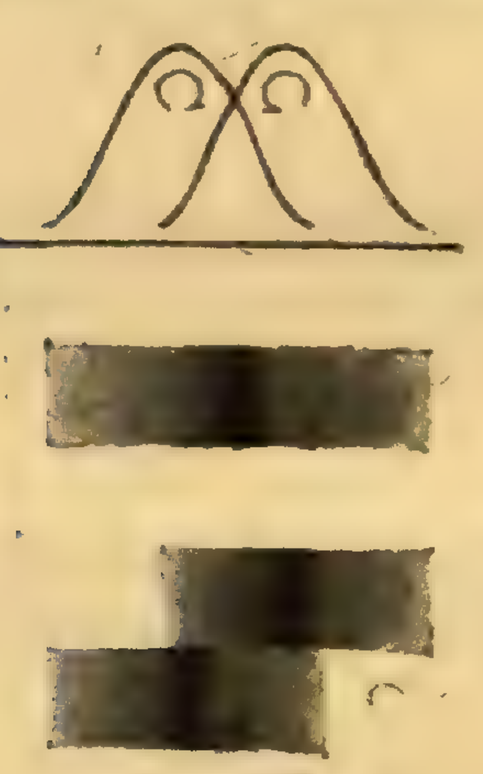

Fig. 4.

As a general rule the distance of the components exceeds that of the lines first considered.

7. Parallel to the lines of force a partial, not too small, overlapping of the components produces a black line limited by two less dark parts. This case is illustrated diagrammatically in Fig. 4.

The two components may be separated by a circular analyser.

These considerations may be applied to the magnetic division in

sun-spot spectra; as a general rule we may expect that the separation of lines in spot spectra becomes more distinct and of larger amount by the use of analysers.

The introduction of a Nicol in the beam may also reveal lines invisible without analyser.

Several peculiarities observed in the distribution of intensity in spot lines, remind one of the now specified superposition phenomena [1]); cf. § 19 below.

8. Superposition effects of nearly, though not exactly, the same nature occur if lines with the same direction of vibration are superposed and if the continuous source of light emits unpolarised light. In the more complicated divisions the now specified superposition occurs also. It is just possible that the superposition of the outer components of the sextet, type D_2, produces only dark, that of the inner and the next outer components, black lines in the continuous spectrum.

It is easily seen that also in the case of the quartet, type D_1, black lines may be produced. The darkest parts may be seen somewhat nearer to the middle of the complete figure, than the outer components of the quartet.

It seems unnecessary to illustrate this by figures. Examples of the specified actions will be given presently.

9. Our observations and spectrograms relate besides to the two principal directions (parallel and at right angles to the lines of force), also to directions inclined to the field.

In the present, first, communication, observations are discussed, relating to 5 different angles between the field and the direction of propagation of the beam (VOIGT's φ, LORENTZ's ϑ).

These values are: 90°, 0°, 60°, 45°, 36°—39°.

The results of the work relating to these angles have been recorded on nearly 100 spectograms.

10. *Observations perpendicular to the field.*

In the upper of the three stripes which are present in the field of view (see § 5), the light vibrates vertically; in the lowest one horizontally, whereas the middle part relates to natural light.

Under the influence of the magnetic field we therefore see the vertically vibrating components as narrow black lines. The quartet of the D_1 line, the sextet of the D_2 line, may be seen very clearly

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[1]) A figure equivalent to the one now given concerning the influence of superposition of magnetically divided components was already drawn for *emission* lines in ZEEMAN. Doublets and Triplets in the Spectrum produced by external magnetic forces. Phil. Mag. July 1897 § 7.

by this method. A small disturbance is produced by the narrow reversed lines due to the electric arc light. The intensity of these lines depends upon somewhat variable circumstances of the arc itself. In some cases these lines are almost invisible, in other ones more prominent. They are to be seen on some of our reproductions ; with our present subject they have nothing to do.

As regards the central stripe we refer to the remark previously made, that the image of the separation must become, on account of the only partial absorption, rather indefinite and weak. (§ 3).

The partial superposition of components gives, at least in the case of diluted vapour, the most conspicuous lines. (§§ 6 and 7).

In the case of the quartet, for example, one sometimes sees instead of four, only two components, situated between the inner and outer ones.

We made experiments with different vapour densities. The observed phenomena may be classified under three phases :

1. The vapour is *very dilute*. The components are clearly visible in the upmost and lowest stripe. In the central stripe the absorption is either hardly perceptible (Plate I, Fig. 1) or the components of the quartet and the sextet are seen as separate, but weak lines. (Plate I, Fig. 2).

In this phase of the phenomenon the great difference of definiteness of the central and outer regions is very remarkable. This contrast is still more marked with eye observation.

In order to obtain good photographs, it was necessary to increase the density of the vapour above the one required for the observation of the very first trace of absorption.

2. Vapour of *intermediate* density.

The components in the upmost and lowest stripes are now no more separately visible or only in the case of the quartet. In the central stripe a superposition of the kind mentioned in § 6 takes place. In place of the quartet an apparent doublet is seen, the components of which are situated between the outer and inner components of the quartet. This case is very clearly represented in Plate I, Fig. 3.

The phenomena exhibited by the sextet (D_2 line) become rather complicated.

The superposition phenomenon is often very distinct. The D_2 line on Plate I, Fig. 3 shows sufficiently the appearance.

3. With still *denser* vapour, the components become very broad and the magnetic change hardly visible. The *polarisation of the edges* of the broad line may be recognized. This phase is represented in Plate I, Fig. 4. It corresponds to the emission effect as it was first discovered : a slight change of broad lines in a weak field.

With still greater absorption the influence of the field becomes imperceptible.

All these phases appear with great regularity. If the intensity of the field is known, it seems possible, the resolving power of the spectroscope being given, to deduce the density of the vapour from the nature of the observed phenomena.

The magnetic division phenomena hitherto observed in sun-spots appear to fall under the second and third phases above mentioned. HALE from measurements of spot lines, compared with laboratory experiments, deduces a maximum intensity of the spot field of 4500 Gauss. Hence, one would be inclined to think that the density in the layers, which bring about the absorption in the sun-spot spectrum can only be small. Moreover, the non-uniformity of the field of sun-spots produces by itself a widening of the components. Light from a limited portion of the spot would give perhaps very narrow spectral lines. In the light, however, of the critical remarks of KAYSER [1]) concerning our knowledge of the influence of pressure and of temperature on spectra all such considerations most be put forward with great diffidence.

11. *Observations parallel to the lines of force.*

In the present experiments the absorbing vapour subjected to magnetic forces is placed between perforated poles.

After putting on the current, one sees in the continuous spectrum, 2 dark bands in the case of D_1, 4 in the case of D_2, according to the diagrammatical figure 1. The absorption is incomplete also now, because of some wave-lengths only right-handed circularly polarized light, but not left-handed is absorbed and the reverse. In order to observe the separation and the polarization a FRESNEL rhomb is placed with its principal plane at an azimuth of 45° with the horizon, a horizontal slit being placed in one of the perforated poles. The FRESNEL rhomb converts circularly polarized into plane polarized light. By means of a calcspar rhomb also now three stripes are obtained. The first phase (very dilute vapour) is represented in Plate I, fig. 5.

Vapour of intermediate density (second phase) exhibits the superposition phenomena mentioned in §§ 7 and 8, and diagrammatically illustrated by Fig. 2. In the central strip *one* line, at the position of the unmodified one, surrounded by feebly absorbing regions, is

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[1]) KAYSER. Handbuch. Kapitel V. Bd. II.

seen. Plate I, Fig. 6 shows these lines for the doublet and the quartet; especially with D_2 the effect is very marked.

12. *Observations in directions inclined to the field.*

According to LORENTZ's elementary theory of magnetic division one generally observes in a direction, which is oblique under an angle ϑ with the lines of force, a triplet with elliptically polarized outer components [1]).

The ellipse, which characterizes the state of polarization of the components with period $T_0 + v$, is the projection on the wave-front of the circle perpendicular to the field, in which the electron with period $T_0 + v$ is moving. v is a small quantity. The direction of the motion of the moving electron also determines the motion in the ellipse. The ratio of the axes is as 1 to $cos\ \vartheta$. For the other outer component with period $T_0 - v$ holds *mutatis mutandis* the same reasoning.

The central line with the unmodified period T_0 always remains linearly polarized. The vibrations of the middle component are in the plane determined by the ray and the line of force and the amplitude of the vibrations is proportional to $sin\ \vartheta$.

If we put $\vartheta = 0$, i. e. in the case of the longitudinal effect, only circular motions remain.

All this applies to very narrow spectral lines in a strong field, the distance of the components being much greater than their width.

According to VOIGT and LORENTZ we must expect some interesting particularities if this restriction be discarded. We return to this point later on.

As a general rule the deductions from the elementary theory are verified. Also in the case of the quartet and the sextet the outer components become elliptically polarized, as has been observed already by RIGHI [2]).

In contradiction with the elementary theory, though not strictly applicable to the case, is the very slight diminution of intensity of the middle components of the quartet even for $\vartheta = 45°$.

13. *Observations at* $\vartheta = 60°$.

If the observation is made with a calcspar rhomb, the image

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[1]) cf. RIGHI l. c.

[2]) RIGHI's observations l. c. all refer to an angle of nearly 55°, the angle at which according to the elementary theory the three components of the triplet are of equal intensity.

remains as with the transversal effect. Yet the presence of elliptic polarization ought to manifest itself by the appearance in the lowest stripe of lines, corresponding to the outer components.

With very dilute vapour and with that of intermediate density as good as no trace of it is seen.

Fig. 7, Plate II shows the first phase with dilute vapour, Fig. 8 the second phase with denser vapour. Only traces of absorption, indicative of elliptic polarization can be seen near D_2, Fig. 8.

The ellipticity is, however, undoubtedly proved by means of the FRESNEL rhomb, placed with its principal plane at an azimuth of 45° with the horizon. Fig. 9 shows the appearance.

The outer components of the quartet towards the red or towards the violet, dependent upon the stripe and the direction of the field, are now considerably weakened; in the case of the sextet they have vanished altogether. All this proves the elliptical polarization of the outer components. For, if the polarisation were linear, as might be inferred from observations with the calcspar alone, then the observation with calcspar and rhomb combined, ought to show no difference between the upmost and lowest stripe. The light of all plane polarized components would issue circularly polarized from the rhomb and, the calcspar making no selection between right-handed and left-handed polarizations, the components towards red and towards violet would all be alike. Such a condition is disproved by photographs such as Fig. 9.

14. One point must be considered somewhat more in detail. What is the reason that the ellipticity is not shown by the calcspar rhomb alone, whereas its existence is most clearly demonstrated by means of the FRESNEL rhomb?

Let an elliptic vibration with vertical axis b, horizontal axis a, be incident upon the rhomb, the principal plane of which is at an azimuth of 45°.

It is easily proved that the elliptic vibration issuing from the FRESNEL rhomb has its axes in the same direction as the original motion and a ratio of the axes $\frac{a_1}{b_1} = \frac{b-a}{b+a}$, the original ratio being $\frac{a}{b}$.

If a be small in relation to b (an elongated ellipse), then, the light issues from the FRESNEL as a more circular vibration, which is more easily analysed.

It depends upon the magnitude of a, whether $\frac{a}{b}$ is *greater* or *less* than $\frac{b-a}{b+a}$.

We distinguish the following cases :

1. a very small, then $\frac{b-a}{b+a} > \frac{a}{b}$.

2. $a = 0{,}414\ b$, then $\frac{b-a}{b+a} = \frac{a}{b}$.

3. $a > 0{,}414\ b$, then $\frac{b-a}{b+a} < \frac{a}{b}$.

We shall apply these results to the interpretation of our observations.

Two cases dependent upon the magnitude of a are of principal importance.

In the first case we can observe the effect of both the axes of the ellipse by means of the combination of the FRESNEL rhomb and the calcspar (*this is the case of the quartet*) (D_1 Fig. 9), whereas without FRESNEL rhomb no effect of the small axis is visible. In the second case the effect of the small axis becomes apparent by the use of the calcspar, whereas its existence cannot be demonstrated with the FRESNEL, the value of $\frac{b-a}{b+a}$ being too small. *This case is represented by the sextet*, (D_2 Fig. 9).

If the observation is made by means of the calcspar rhomb, we indeed see with dense vapour new components in the lowest stripe (see Fig. 8, D_2). The theoretical import of this result will be discussed on another occasion.

After introduction of the FRESNEL rhomb the component to the left of the central line (small axis of the ellipse) remains invisible. (Fig 9, D_2, inferior stripe).

Hence we may conclude that at the angle now investigated the ellipticity of the outer components of the *sextet* (the ratio $\frac{a}{b}$) exceeds that of the quartet (and is also larger than 0,414).

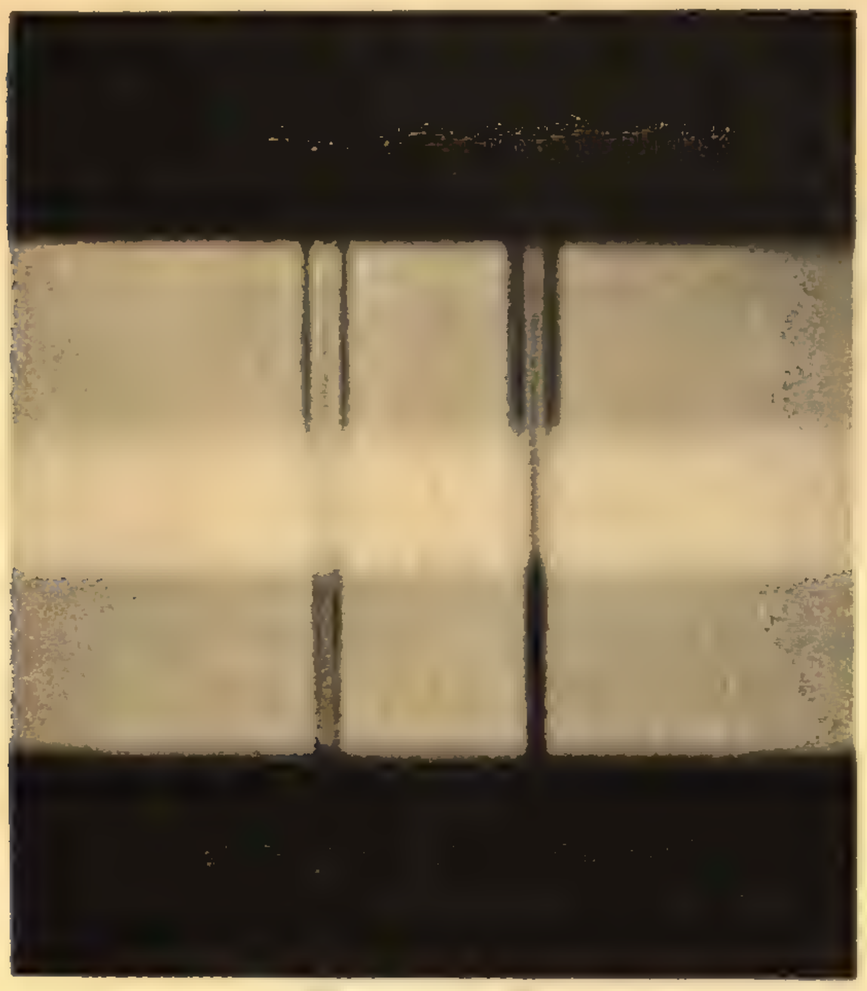
15. *Observations at* $\vartheta = 45°$.

The photographs taken with the calcspar alone, show very clearly the ellipticity of the outer components.

With vapour of intermediate density the phenomenon is already very marked, especially in the case of D_2 (Plate II, Fig. 10). Very remarkable is the slight diminution of intensity of the inner components of the quartet. According to the elementary theory the intensity of the central component of *a triplet* ought to have diminished already to less than *half* the original value.

Prof. P. Zeeman and **Dr. B. Winawer.** The magnetic separation of absorption lines in connexion with sun-spot spectra.

1.

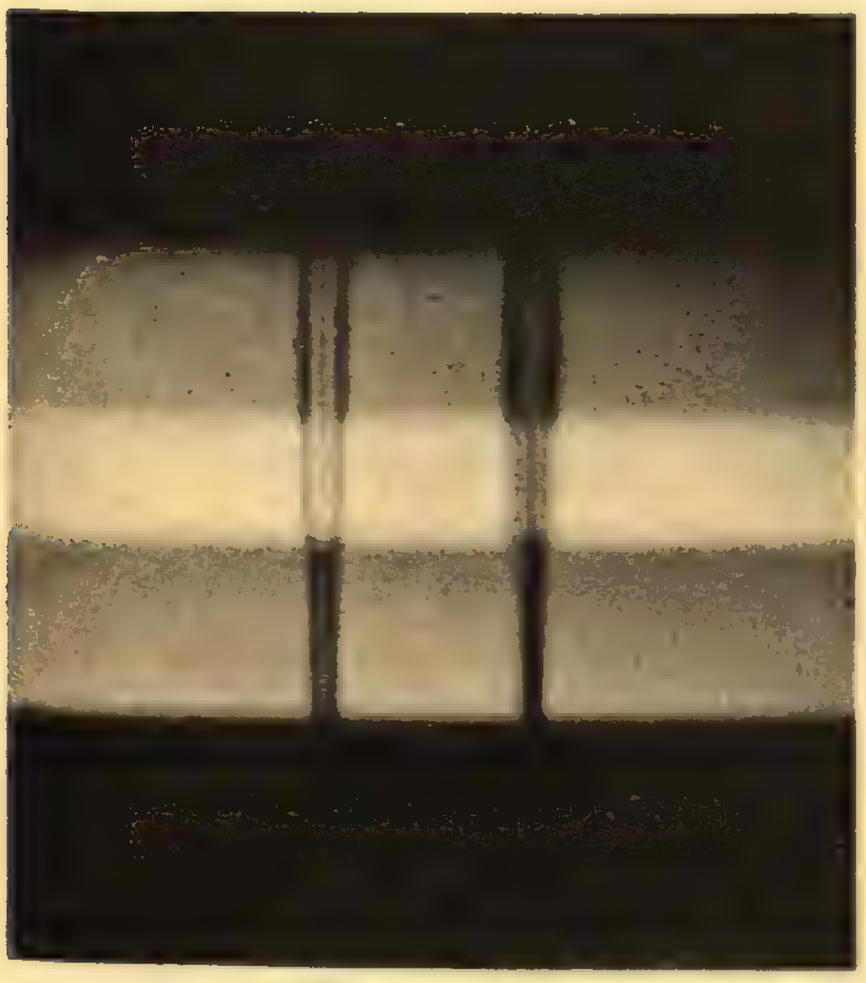


D_1 D_2

3.

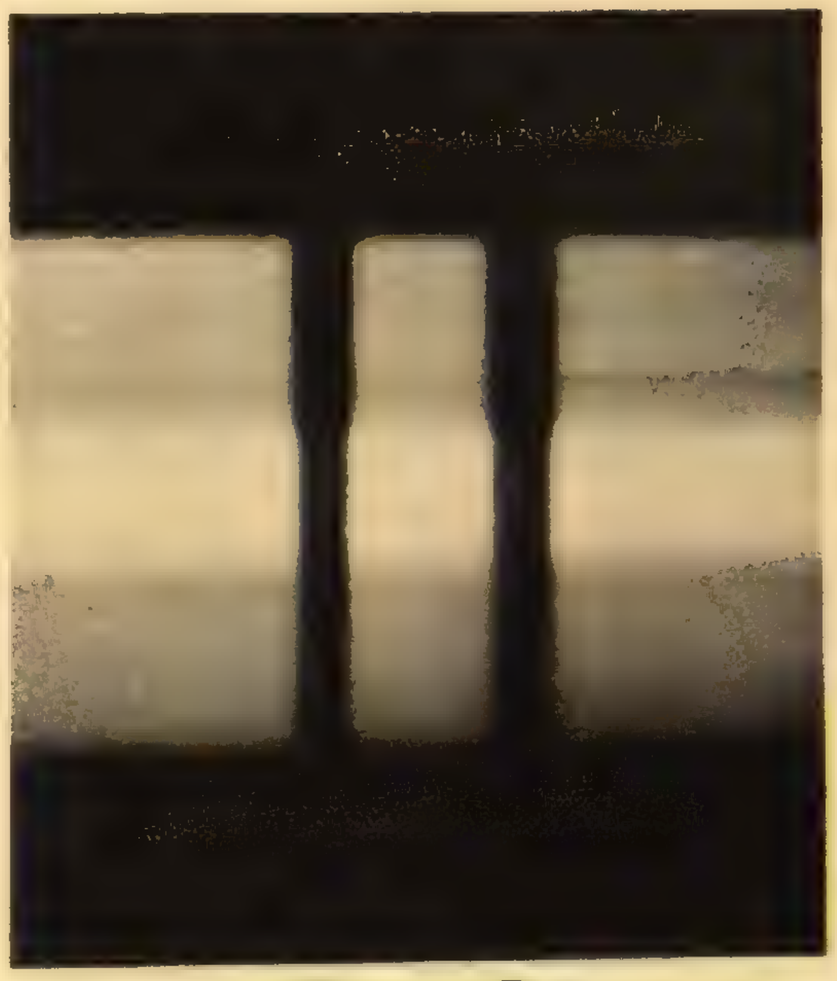
D_1 D_2

2.



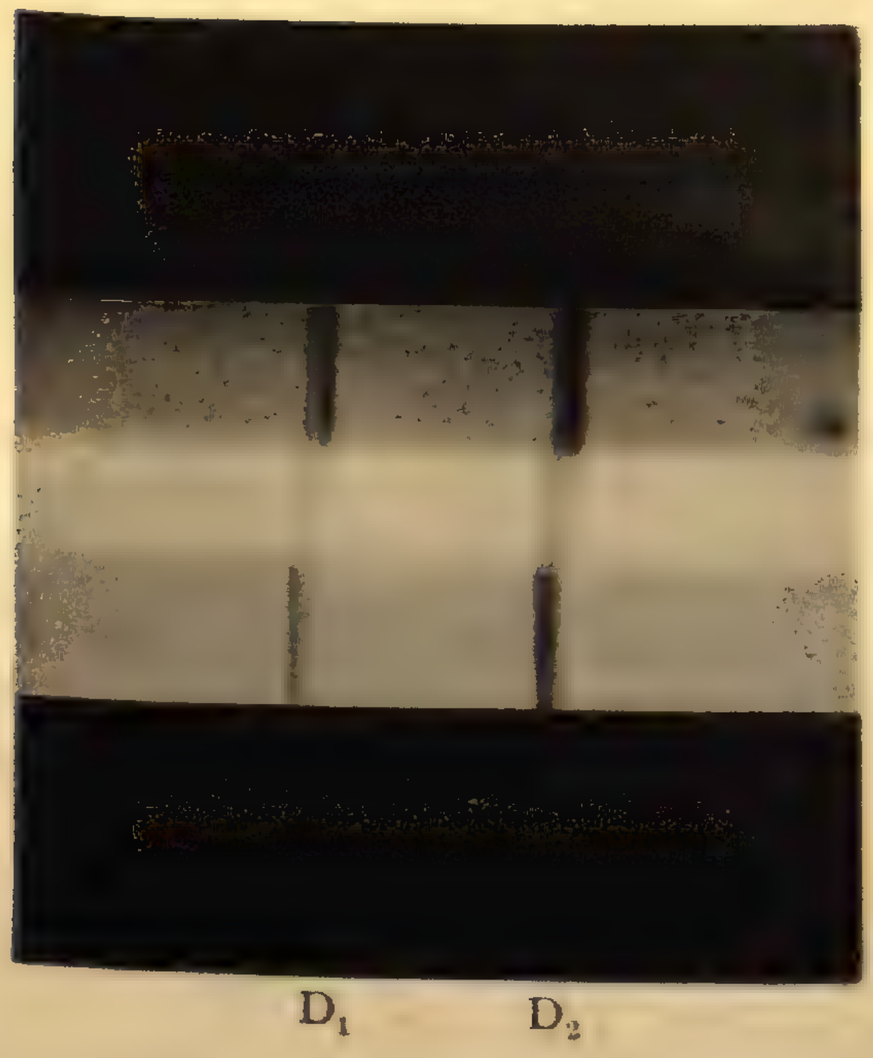
D_1 D_2

4.



D_1 D_2

5.



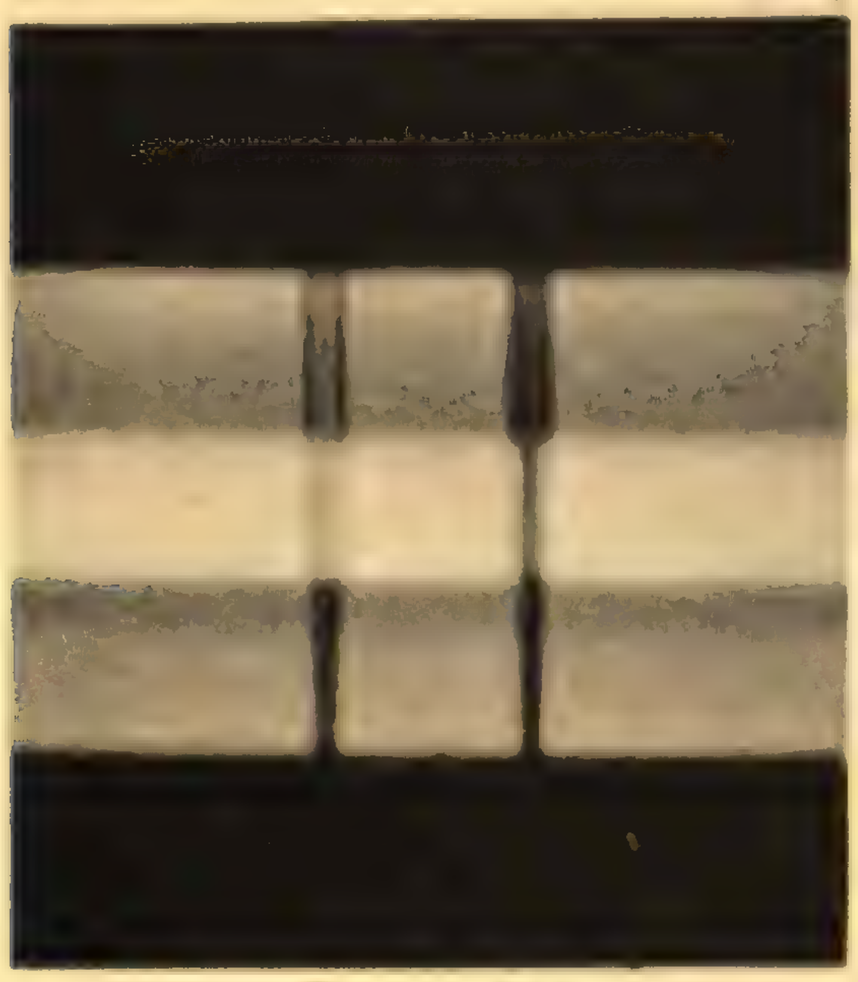
D_1 D_2

6.

D_1 D_2

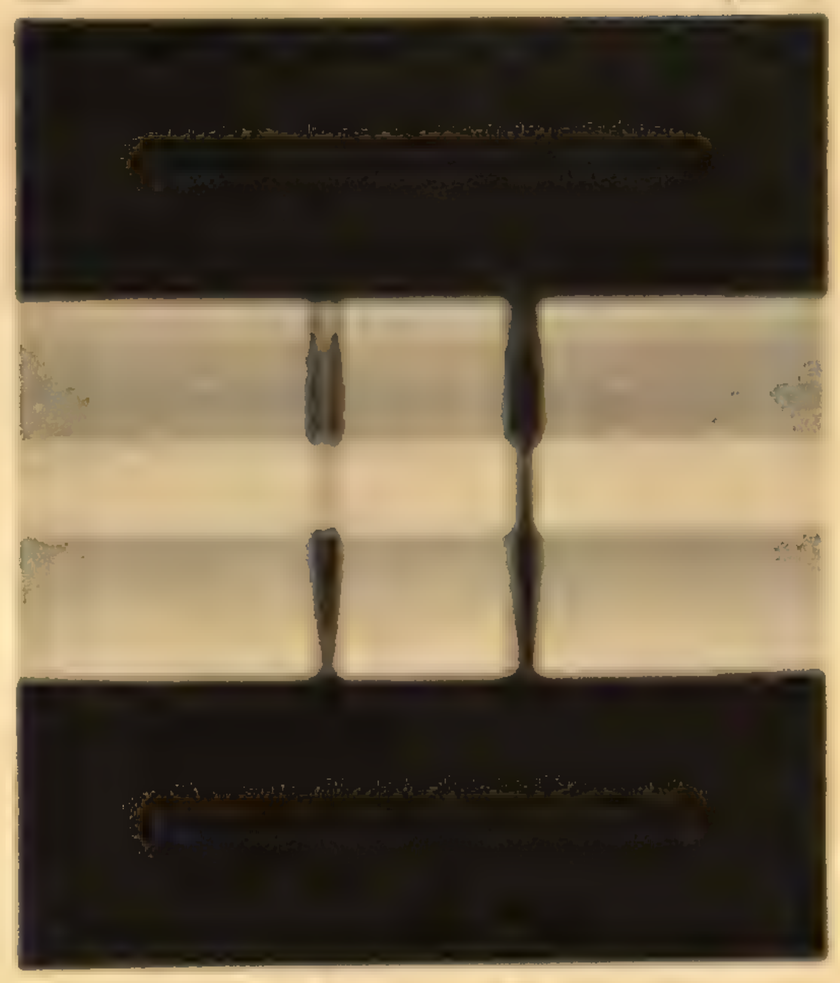
Prof. P. Zeeman and Dr. B. Winawer. The magnetic separation of absorption lines in connexion with sun-spot spectra.

7. (60°)



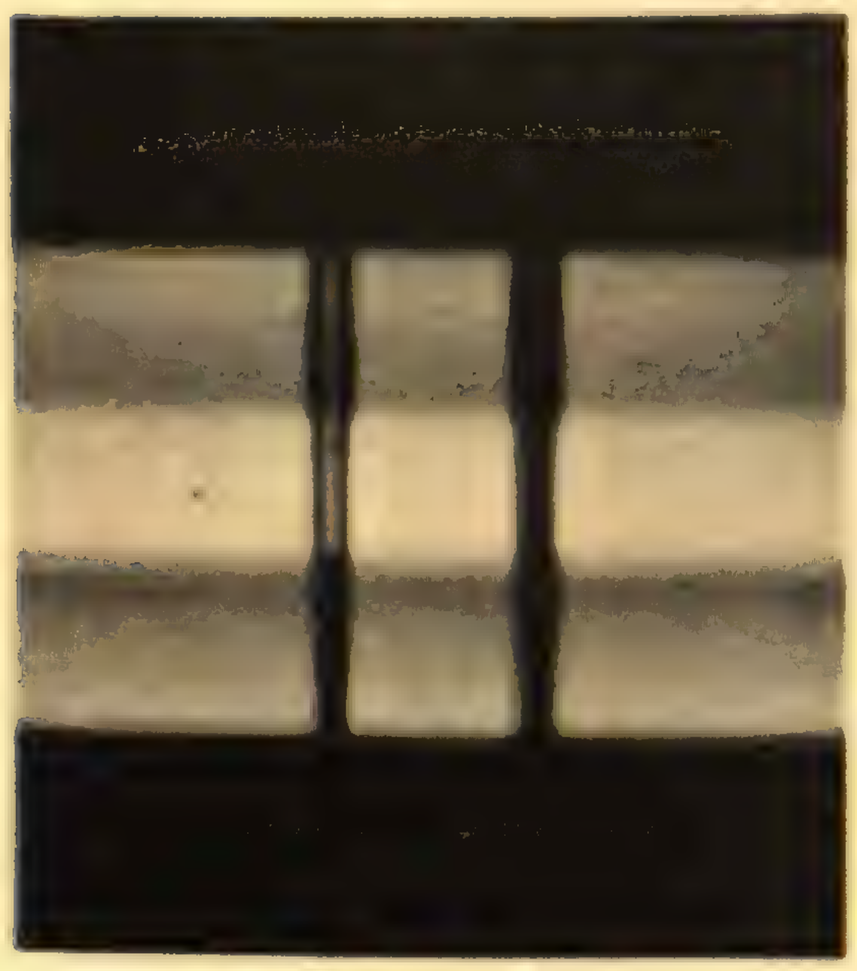
D_1 D_2

10. (46°)



D_1 D_2

8. (80°)

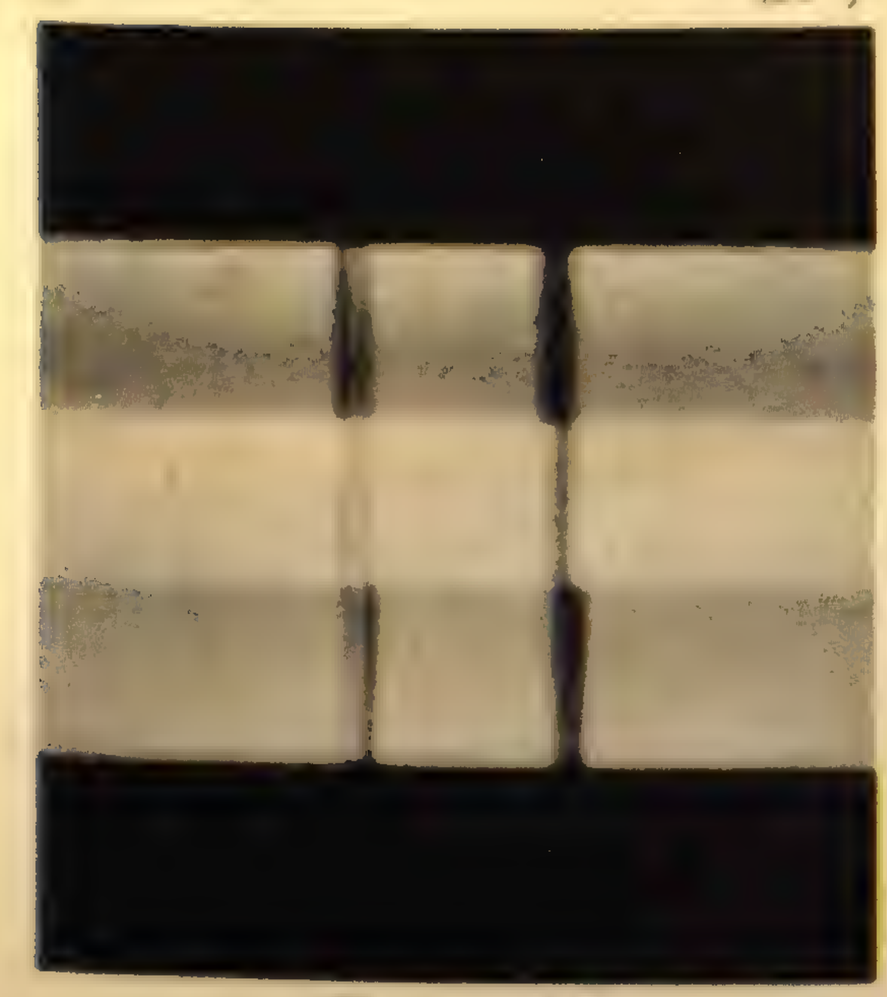


D_1 D_2

11. (45°)

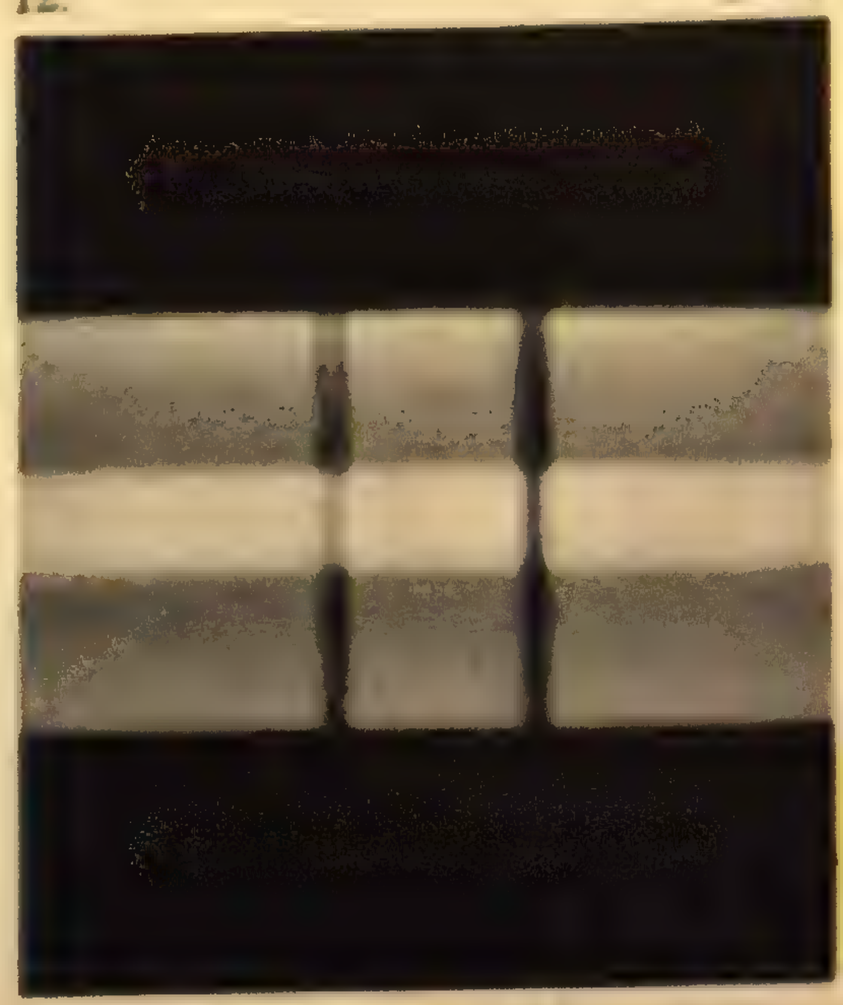
D_1 D_2

9. (60°)



D_1 D_2

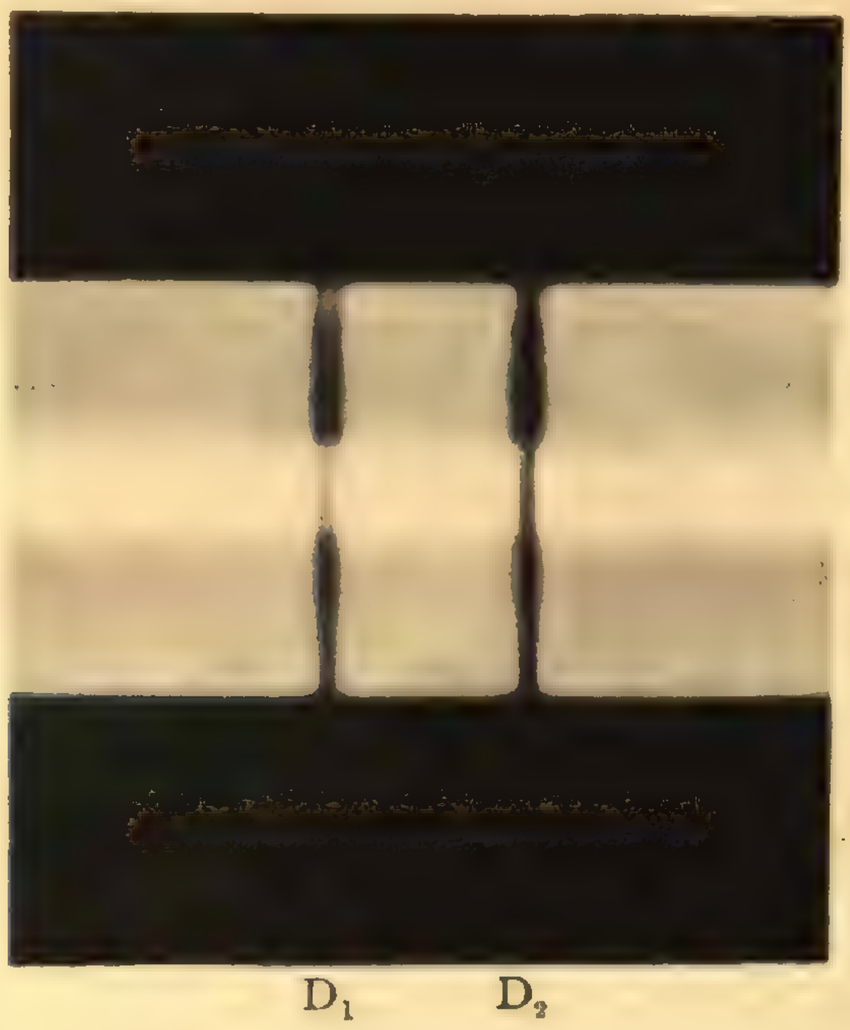
12. (39°)



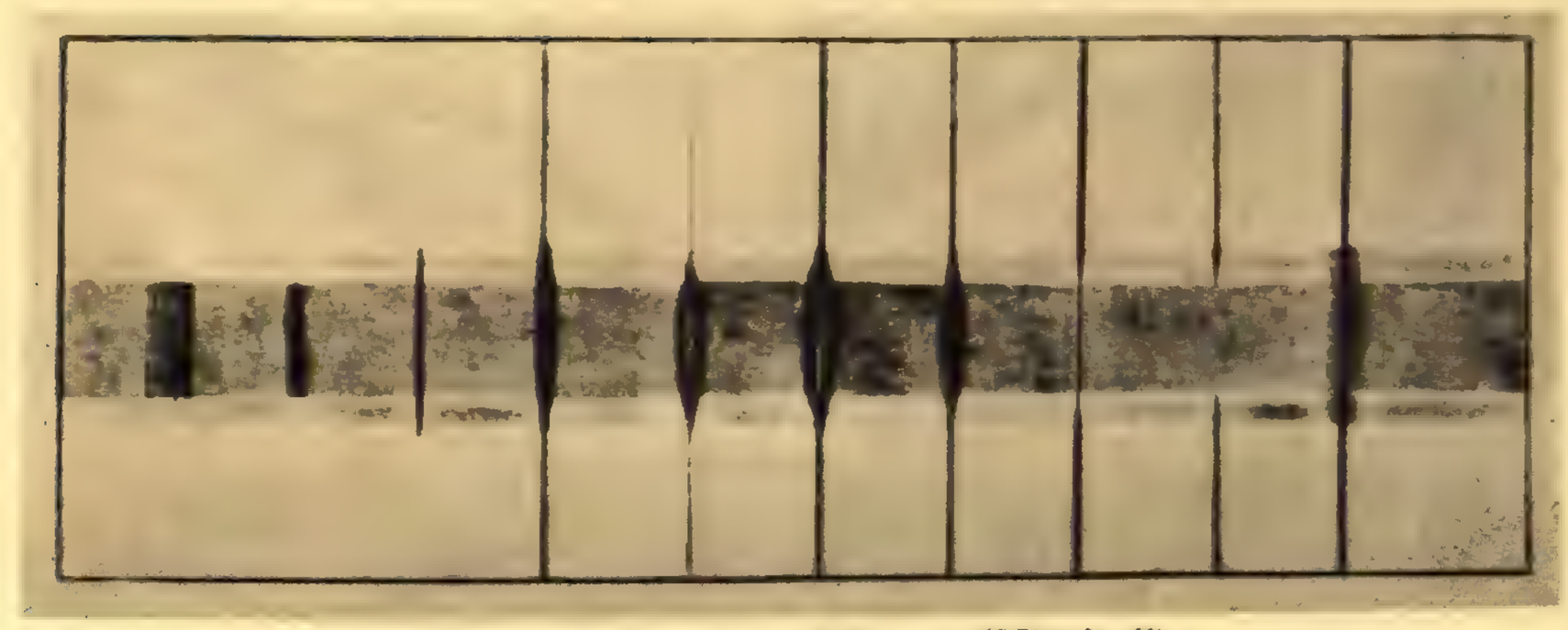
D_1 D_2

Prof. P. Zeeman and **Dr. B. Winawer.** The magnetic separation of absorption lines in connexion with sun-spot spectra.

13. (39 °)



14. 1 2 3 4 5 6 7 8 9 10

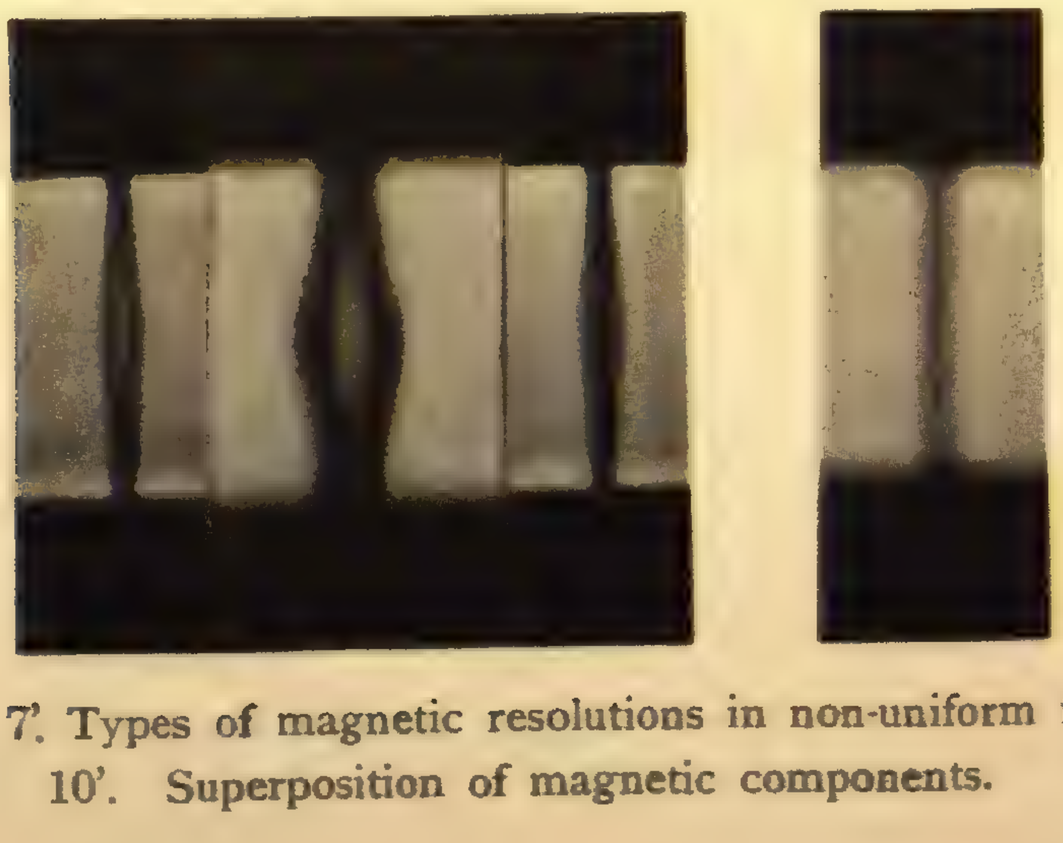


Types of sun-spot lines. (Mitchell)

5, 6. Widened lines with centres reserved bright.

7. Widened and weakened line. 10. Winged line.

15. 5^1 6^1 7^1 10^1



5', 6', 7'. Types of magnetic resolutions in non-uniform fields.

10'. Superposition of magnetic components.

16. If a FRESNEL rhomb combined with a calcspar rhomb is introduced in the beam, one of the components of the quartet also entirely disappears. At an angle of 60° this was only the case with the sextet. (Plate II, Fig. 11).

17. *Observations at* $\vartheta = 39°$.

The elliptic polarisation tested by means of the calcspar rhomb is very marked, even with dilute vapour (Plate II, Fig.12, Plate III, Fig.13).

The inner components of the quartet are now decidedly less intense than the outer ones.

Plate III, Fig. 13 especially shows the smaller intensity of the components of D_1 in the lowest stripe. Indeed, they are unmistakably thinner than those in the upmost stripe.

18. According as the angle between the ray and the lines of force is diminished, the intensity of the field must diminish at the same time. In order to make it possible for the rays to traverse the field under smaller angles the vertex semiangle of the cones must deviate more and more from the theoretical optimum of nearly 55°.

The decrease of the magnetic separation is clearly shown in our photographs.

We intend to communicate on another occasion experiments under smaller angles ϑ and to enter upon some details concerning the case in which the components of the triplet are not neatly separated. Some measurements of the ellipticity of the components will also be given. On the present occasion we only intended to give a general survey of the inverse effect, illustrating it by some particular cases.

19. *Types of separation in spot and laboratory.*

In one direction we shall now enter upon some more details. The magnetic separation of lines in a *non-uniform* field has been treated on a former occasion. [1]) The results then obtained and our present observations may be of some interest in connection with certain phenomena observed by HALE. We intend to return to this subject. Presently it seems interesting to allude to MITCHELL's descriptions of the various types of spot lines as indicated in the diagram published in the Transactions of the International Solar Union [2]).

Our Fig. 14, Plate III has been copied from this source. The types 5, 6, 7, and 10 of the Figure are very characteristic. Type 9 perhaps falls under the type of lines invisible without Nicol mentioned

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[1]) ZEEMAN. These Proceedings, April 1906, November 1907.

[2]) Transactions Intern. Union Solar Research, p. 199 etc. 1908.

§ 7 above. In Fig. 15 are represented some separations observed in the laboratory *without* Nicol or other analyzer, 5′, 6′, 7′ have been taken in non-uniform fields. 5′ is the quartet of D_1 observed across the field; 6′ the sextet of D_2 observed axially in a non-uniform, in the central part very strong, field; 7′ also refers to D_2 in a weaker field, the observation being made across the lines of force. The type 10′ refers to the D_2 line, when observed in a direction parallel to the field. The field is uniform. The separation gives an example of the superposition phenomenon mentioned in § 7.

The analogy of the type 10′, Fig. 15 and the type of the "winged line" seems very remarkable. Of course observation of the state of polarization would be necessary in order to prove the analogy.

EXPLANATION OF PLATES I—III.

The figures 1—13 are about thirteenfold enlargements of the images given by the grating of the absorption lines D_1 and D_2 in a magnetic field.

The upmost and lowest of the three stripes of these figures relate to (oppositely) polarized light; in the central part the phenomenon is represented as it is seen in natural light.

PLATE I. 1, 2, 3, 4, observations $\perp$ lines of force with different vapour density.
5, 6, observation // lines of force with different vapour density.

PLATE II. 7, 8, observation at $\vartheta = 60°$ calcspar rhomb alone.
9, $\vartheta = 60°$, calcspar combined with FRESNEL rhomb.
10, 11, $\vartheta = 45°$.
12, $\vartheta = 39°$.

PLATE III. 13, $\vartheta = 39°$.
14, Types of sun-spot lines (adopted from MITCHELL).
15, 5′, 6′ 7′, separations in non-uniform laboratory fields. 10′ superposition phenomenon § 7.

Physics. — "*The thermomagnetic properties of elements.*" By Prof. H. E. J. G. DU BOIS and Prof. KŌTARŌ HONDA. (Communication from the Bosscha-Laboratory).

(Communicated in the meeting of January 29, 1910.)

In 1895 CURIE[1]), though he had investigated relatively few substances, believed that he could formulate his results in the following rules:

1. For paramagnetic substances the specific susceptibility is inversely proportional to the absolute temperature.

2. For diamagnetic substances, on the contrary, the susceptibility is almost independent of temperature.

3. For the latter class of substances, changes of physical state generally have hardly any influence.

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[1]) P. CURIE, Ann de Chim. et de Phys. (7) **5** p. 289. 1895. – Oeuvres p. 232 Paris 1908.

4. The same holds for variations of chemical state (allotropy).

One of us in 1900 proposed to call the first of these thermomagnetic rules CURIE's law and to introduce a *Curie's constant* such that:

$$\chi(\theta + 273) = C.$$

It was also expressly stated that very probably this was only a kind of "limit-law" in the sense of the analogous law for ideal gases. In addition it was very soon shown that the usual theory of directed magnecules leads to such a law, when generalised from a more magnetokinetic point of view; this was theoretically proved and experimentally confirmed in the LORENTZ- and BOSSCHA-volumes of the "Archives" [1]). With all due regard for CURIE's important researches and for his first rule, the second can and could have no general signification, for it at once contradicted the results of other observers, e. g. as in the special case of water.

With regard to the third and fourth rules even their author pointed out several exceptions. As the values of the susceptibilities of the diamagnetic substances tested proved much less than those of the paramagnetic bodies, CURIE came to the conclusion that these two opposite forms of magnetic induction were due to completely different causes.

Starting from these experimental conclusions, LANGEVIN [2]) in the year 1905 elaborated an electronic theory of magnetism; he also gave a kinetic representation of CURIE's first law, completely analogous to the one mentioned above, without, however, mentioning it, and which is in addition perfectly independent of the introduction of electrons.

It appeared, therefore, desirable to investigate the thermomagnetic properties of more substances: in the first place those of elements, in order to judge whether CURIE's conclusions admit of such an extensive generalisation. It may be at once remarked that such is not at all the case.

Experimental Arrangement. The method, previously used by CURIE and other investigators, of the torsion-balance combined with a non-uniform field was applied, employing the semicircular electromagnet recentiy described in these Proceedings. The axes of the two cores formed an angle of 10° to 20°; the maximum gradient of the field then lies at a certain distance to one side of their point of intersection. The field

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[1]) H. DU BOIS, Rapp. Congr. d. Phys. **2** p. 486, Paris 1900. — Arch. Neérl. (2) **5** p. 246, 1900, also **6**. p. 581, 1901. — Verh. nat. en gen. Congr. **8** p. 60, Rotterdam 1901. *Notations*:

a, Atomic weight.	θ, Temperature.
C, CURIE's constant.	χ, Specific susceptibility.

[2]) P. LANGEVIN, Ann. de Chim. et de Phys. (8) **5** p. 70, 1905. Journ. de Phys. (4) **4** p. 678, 1905.

itself at this particular point amounted to 25 kilogausses; it was measured from point to point by means of a small standardised spherical test-coil. The sensitiveness of the torsion-balance could be varied; it was determined in the usual way by means of applied additional moments of inertia.

The furnace consisted of a porcelain tube wound with platinum wire and insulated with kaolin powder and asbestos. With a consumption of 1.2 kilowatts a temperature of 1250° was attained, which was measured by means of a thermo-element, previously checked by observations on the melting-points of tin, tellurium, antimony, and gold.

Test-samples. The great difficulty with all experiments in this sphere of work is and always will be the prevalence of iron, with its overwhelming ferromagnetic properties, though it hardly ever seems to act quite freely. In the case of fifteen elements, their binary alloys with iron were examined in TAMMANN's laboratory, not in the very diluted state, however, which generally corresponds to ferrugineous impurities. Of 81 elements, 43 were tested; many of them were supplied as pure as possible by KAHLBAUM; Prof. COHEN and Dr. HOITSEMA of Utrecht kindly placed several elements at our disposal; as yet the 10 gaseous elements have not been tested; Li, Rb, Cs, Ca, Sr, Ba could not be obtained sufficiently free of iron; while Be, Sc, Ga, Ge, Y, Rd, and the rare metals could not be procured. Fe, Co, Ni, of course, form a class by themselves. Dr. M. HANDA kindly determined the percentage of iron colorimetrically by the Berlin blue-reaction.

The experimental results, moreover, furnish certain physical criteria of their own reliability, for in so far as the susceptibility proves independent of the field there can hardly be question of a ferromagnetic ingredient. With about one third of the samples this was not the case, for the susceptibility diminished (in the algebraic sense) with an increasing field according to a hyperbolic law. From this Mr. MORRIS OWEN calculated the value χ_∞ which would hold asymptotically for an infinite field; and, in addition, the influence of the ferromagnetic ingredient, which at most amounted to only one sixth — and generally much less even — of what could be imputed to the iron in the free state. The thermomagnetic properties also afford a test of purity up to a certain point; a few strongly ferrugineous substances show a great diminution of susceptibility between 500° and 600°, whilst above 700° the influence of iron hardly need be feared. In no case is there reason to doubt that the value of the susceptibility of absolutely non-ferrugineous elements would remain constant, at least within the usual

field-range. The full communication of the results obtained would require many tables and curves; we therefore draw attention to the principal points only.

Specific susceptibility [1]) *at* 18°. The values found lie between — 2 and + 5 (amorphous carbon and palladium respectively). It cannot be maintained that the positive paramagnetic values are on the whole larger than the negative diamagnetic ones. Oxygen alone forms an exception with a value of about 100; the value for manganese was approximately 10; this contained, however, $^1/_3{}^0/_0$ of iron.

CURIE had already pointed out the influence of allotropy in the case of phosphorus and antimony, and also that there is no such influence with sulphur, though it is so well-known for its polymorphous properties. A difference was shown to exist between diamond (— 0,49) and amorphous carbon (— 2,02); silicium crystalline (0,12) and amorphous (— 0,14); and especially between common tetragonal tin (+ 0,03) and grey tin (— 0,29). In the case of tin, the first — the tetragonal — was KAHLBAUM's very pure electrolytic material; it was afterwards inoculated with a small quantity of grey tinpest, kindly sent by Prof. COHEN from the stores of the VAN 'T HOFF Laboratory.

For weak fields indium seemed to be paramagnetic; in a field of 7 Kgs. the value of the susceptibility passed through zero and became negative, doubtless in consequence of $0{,}013{}^0/_0$ iron: this phenomenon is of no consequence because it is also discovered in complicated substances such as certain kinds of porcelain, glass, etc.

Notwithstanding many omissions, it was still possible to follow the general course of the curve $\chi =$ funct. (a); the curve appears to be rather intricate, but still shows a distinct relation to the periodic system. According to the arrangement of MENDELEJEFF-BRAUNER's table, the rows (1, 2, 3, 4), (5, 6, 7, 8), and (9, 10, 11, 12) each form a division I, II, III in which the shape of the curve repeats itself in a peculiar way. At the junction of I and II Cr, Mn, Fe, Co, Ni lie on a positive maximum; between II and III, in the same way, the "rare" metals: within I, II and III the diamagnetic negative peaks are occupied by the similar pentavalent elements P, Sb and Bi of the fifth group (3^{d}, 7^{th}, 11^{th} row). In more than one respect further magnetic analogies of secondary importance exist, which, however, must be left unmentioned in this communication.

Susceptibility at high temperatures. As a rule the path of the curve $\chi =$ funct. (θ) for any substance proved to be the same when the temperature was increased or afterwards decreased; certain deviations probably depend on a change of condition of the iron present after

[1]) Everywhere below expressed in millionths.

PARAMAGNETIC			DIAMAGNETIC		
C	*I*	*D*	*C*	*I*	*D*
Na (0°—97°)	Ti	[O]	B, cryst (400°—1200°)	B cryst (0°—400°)	C, amorphous
Al (657°—1100)°	V (500°—1200°)	Mg (0°—657°)	Si, crystalline	Diamond	Cu
K (0°—150°)	Cr (500°—1100°)	[Fe] (800°—1350°)	P, white	Ag	Zn (300°—700°)
V (0°—500°)	Mn	Co (1150°—?) ?	S, rhombic	I (0°—114°)	Zr (500°—1200°)
Cr (0°—500°)	Mo	[Ni] (350°—800°)	Zn (0°—300°)		Cd (300°—700°)
Nb (0°—400°)	Ru (550°—1200°)	Pd	As, Sublim.		In (0°—150°)
W	Rh	Rare metals?	Se, Metallic		Sb (0°—631°)
Os	Ir	Ta	Zr, cryst (0°—500°)		Te
	Th (0°—400°)	Pt	Cd (0°—300°)		I (114°—200°)
		U	Au		Tl
			Hg (0°—350°)		Pb (0°—327°)
			Pb (327°—600°)		Bi (0°—268°)

heating. Mg and Ru shewed the above mentioned diminution in a marked manner between 500° and 600°. The results are collected in the table p. 600. The elements in square brackets have previously been examined by others; the atomic weights in each column increase from top to bottom; the elements under column *C* show a constant susceptibility, under *I* a numeric increase on heating and under *D* a numeric decrease. The fewest number (4) of elements appears in the fifth column, in the case of which the susceptibility increases on heating, the increase being, however, very small in each instance.

From a thermomagnetic point of view a certain relation also exists in connection with the periodic curve $\chi =$ funct. (a): the paramagnetic elements under *D* all lie at the principal maxima or at the secondary peaks; on the contrary, those under *I* lie on the ascending branches of the curve. Therefore the sharpness of the bends would be flattened more and more at higher temperatures; probably at lower temperatures they would become more accentuated, and it may be that only then do they attain their most characteristic shape; of course the temperature of $+18°$ is quite arbitrary. Concerning CURIE's rules the following statements may be made:

1. Only palladium foil from KAHLBAUM, with 0,70% of iron and $\chi = +\ 6{,}12$, on heating followed, more or less, CURIE's law, but on cooling it shewed complications. With much purer palladium from Dr. HERAEUS, with 0,03% iron and $\chi = +\ 5{,}79$, the susceptibility fell less rapidly than would follow from CURIE's rule; temperature-hysteresis was not observed on cooling [1]).

2. There are only 6 diamagnetic elements which do not vary within the whole temperature-range. Of these P, S and Se had already been experimented upon by CURIE.

3. On melting or solidifying, sometimes — not always — a discontinuity appears, which can be classified under one or other of the two following divisions: I, a large or small leap in the curve of χ itself, as with P (44°), Ag (961°), Sn (233°), Sb (631°), Te (450°), Au (1064°), Tl (290°), Pb (327°), Bi (268°); II, a rather sudden change of $d\chi/d\theta$ as with Mg (633°), Cu (1065°), Cd (322°), I (114°); with regard to sulphur, the curve at the melting-point departs slightly from its otherwise absolutely rectilineal character, which variation was probably overseen by CURIE.

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1) By chance palladium is the only paramagnetic element examined by CURIE; perhaps it was not pure enough. The important results for oxygen, for ferromagnetic metals at very high temperatures and also for their salts crystallised or in solution, of course continue to hold.

4. As regards the thermomagnetic examination of polymorphous transformations, a discontinuous diminution of 15% of the specific susceptibility was found at the transition-point of α-thallium and β-thallium at 234°. But the most remarkable properties are shewn by tin: If diamagnetic grey tin is slowly heated, at 32° the specific susceptibility (— 0,29) changes almost suddenly (like the density) and at 35° passes through zero. Possibly this change would wholly take place at the point of transformation (18°) but then at a much slower rate. Further heating continuously increased the susceptibility so that at about 50° the value (+ 0,03) for paramagnetic tetragonal tin was reached, which afterwards remained practically constant; according to DEGENS the point of transformation tetragonal $\rightleftarrows$ rhombic tin lies at 161° at which temperature nothing particular was noticed; at the melting point (233°) a discontinuity from $\chi = +0{,}03$ to $\chi = -0{,}04$ once more appeared; the diamagnetic liquid metal remained nearly unchanged.

An extension of these thermomagnetic investigations towards low temperatures is in preparation.

From the above, especially from the conclusions arrived at under 1 to 4, it seems to follow that CURIE's four corresponding rules are certainly devoid of the general meaning, which has rather rashly been ascribed to them. At the same time the experimental starting-points of LANGEVIN's theory are evidently undermined; more solid and broad foundations for future theories can only be laid with the aid of more extensive research.

Chemistry. — "*Studies on Tellurium*: 1. *The mutual behaviour of the elements sulphur and tellurium*". By Prof. F. M. JAEGER. (Communicated by Prof. VAN ROMBURGH).

(Communicated in the meeting of January 29, 1910).

§ 1. Whilst we are in the main thoroughly informed as to the relation of selenium and sulphur, the views as to the mutual behaviour of the elements tellurium and sulphur still differ somewhat. KLAPROTH [1]) has already investigated this subject. He states that on melting together tellurium and sulphur leaden coloured masses are formed crystallising in rays, which, on heating, give off sulphur and yield a porous metallic looking mass, which he takes to be telluriumsulphide. BERZELIUS [2]), thirty years later again broached the subject; he found that no compounds were formed on melting, but thought that the

[1]) KLAPROTH, Crelle's Ann. (1798). 191.

[2]) BERZELIUS, Gilb.-Pogg. Ann. **8**. (1826). 413.

compounds TeS_2 and TeS_3 are present in the brownish-black precipitates, formed when passing H_2S through solutions of tellurites and tellurates. He arrived at that conclusion on account of the solubility of these precipitates in aqueous potassium or sodium hydroxide, which is also the case with TeO_2 and TeO_3.

BECKER [1]) was the first to analyze these precipitates and he finally arrived at the conclusion that their composition actually corresponds with TeS_2 and TeS_3. He proved however, that nearly all the sulphur may be removed from these substances by treatment with carbon disulphide: TeS_3 yielded a residue containing 6.14 % of sulphur instead of 42.85 %, TeS_2 a residue containing 3.69 % instead of 33.4 %. He concludes that the black precipitates are only mixtures whose composition agrees nearly with those of the supposed compounds. According to him they are formed primarily as ephemeral compounds, which are at once decomposed by the solvent. BERZELIUS [2]) and OPPENHEIM [3]) obtained double sulphides to which they assigned the formulae $3K_2S + TeS_2$, etc. In more recent times, BRAUNER [4]) and GUTBIER [5]) again inclined to the opinion that we are dealing here with mixtures of the elements.

§ 2. Since DUMAS placed tellurium in the sulphurgroup, as the first homologue of selenium, and thus the well-known difficulty as to the position of tellurium, in regard to iodine, in the periodic system introduced later, was created, — the question as to the relation of tellurium on the one side and sulphur and selenium on the other has again become of actual importance. For now it is undoubtedly certain that the atomic weight of tellurium is 127.6 and therefore *greater* than that of iodine. On the other hand the differences between tellurium and the other two elements are so strongly pronounced that RETGERS on account of the isomorphism between tellurates and osmiates, thought it would be better to include tellurium in the group of the platinum metals. Tellurates to wit, are not isomorphous with sulphates, selenates, manganates, ferrates etc. On the contrary, PELLINI showed an isodimorphism in the case of $(C_6H_5)_2SeBr_2$ and $(C_6H_5)_2TeBr_2$, whilst NORRIS and MOMMERS noticed a direct isomorphism between the selenium- and tellurium double chlorides and bromides of dimethyl-

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[1]) BECKER, Lieb. Ann. d. Chem. **180.** (1876). 257.

[2]) BERZELIUS, Traité de Chimie. (1830).

[3]) OPPENHEIM, Journ. f. prakt. Chem. **71.** (1857). 270

[4]) BRAUNER, Journ. Chem. Soc. **67.** (1895). 527.

[5]) GUTBIER, Berl. Ber. **34.** 2114. (1901).

amine. But on the other hand many objections have been raised to the position assigned to tellurium: for instance, the different constitution of telluric acid which, probably, must be looked upon as H_6TeO_6 and the totally different hydration of tellurates in comparison with sulphates and selenates. However this may be, it is highly desirable to obtain more *data* as to the position of tellurium among the other elements and for this reason, the relation to sulphur had to be ascertained in the first place.

§ 3. The tellurium was obtained from $1^1/_2$ kilo of crude tellurium probably derived from American ore. It appeared to contain the following elements: tellurium, selenium, sulphur, lead, copper, bismuth, iron, silicon and traces of antimony, zinc and a few other metals.

The first purification was carried out by oxidation with *aqua regia*, evaporation of the filtrate to dryness, and repeated extraction of the residue with strong hydrochloric acid. The boiling filtrate was then precipitated by sulphur-dioxide; the first portions of the precipitate being rich in selenium were each time rejected. This operation was repeated three times. The amorphous tellurium was divided into two parts; one portion was converted, by the process given by NORRIS, FAY and EDGERLEY [1]), into basic tellurium nitrate $Te_2O_3(OH)(NO_3)$ and by repeating the process five times, which operation lasted many weeks, it was finally obtained quite pure in the form of the said salt: from this, pure TeO_2 was then obtained by gentle ignition and this, dissolved in pure hydrochloric acid was precipitated by SO_2. The other portion was converted into telluric acid by means of CrO_3, according to STAUDENMAYER's process as modified by GUTBIER [2]); this was purified by precipitating twelve times with nitric acid and then crystallising from water. It is necessary to reduce the adhering CrO_3 with alcohol, otherwise the telluric acid crystals retain a yellow colour which is caused by occluded solid CrO_3; this matter I hope to refer to shortly.

The pure telluric acid was then reduced completely by hydrazine hydrate.

The crystalline form of the basic nitrate has not been described up to the present. The following *data* have been obtained from the substance crystallised from nitric acid.

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[1]) NORRIS, FAY and EDGERLEY, Americ. Chem. Journ. **23**. 105.

[2]) GUTBIER, Z. f. anorg. Chem. **29**. 22. (1901); **32**. 96. (1902).

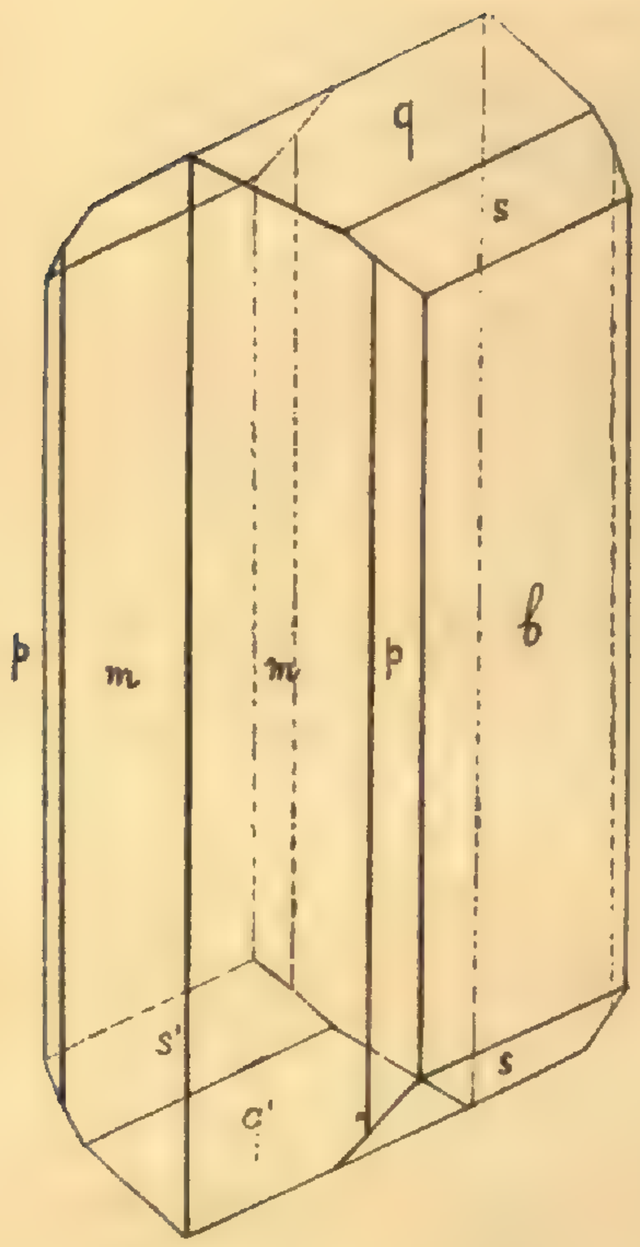

Fig. 1.
Crystalline form of basic tellurium nitrate.

Colourless, very lustrous needles up to 5 m.m. in length and usually flattened along {010}. They exhibit many vicinal planes particularly in the vertical zone, and greater angular differences occur also in different individual crystals. The measurements must, therefore, be regarded only as approximations.

Rhombic-bipyramidal.

$$a : b : c = 0.590 : 1 : 0.607.$$

Forms: $m = \{110\}$, $b = \{010\}$ and $p = \{120\}$, very lustrous; particulary b, which is also a cleavage plane and possesses a high lustre. On the other hand $q = \{011\}$ and $s = \{021\}$ are dull, the form {021} is mostly absent. The crystals exhibit a pronounced inclination to tetragonal symmetry.

Angular values:		Measured:		Calculated:	
$m : m$	$= (110) : (1\bar{1}0) =$	* 61°	5′	—	
$b : q$	$= (010) : (011) =$	* 58	44½	—	
$m : p$	$= (110) : (120) =$	19	25	19°	18½′
$p : b$	$= (120) : (010) =$	39	59	40	2½
$q : q$	$= (011) : (0\bar{1}1) =$	62	50	62	31
$m : q$	$= (110) : (011) =$	74	54	74	43
$m : b$	$= (110) : (010) =$	59	27½	59	27½
$b : s$	$= (010) : (021) =$	39	46	39	29
$s : q$	$= (021) : (011) =$	19	0	19	16

Completely cleavable towards {010}.

The optical axial plane is {001} with the a-axis as first diagonal. Strong rhombic dispersion with $\varrho < v$; the apparent axial angle in cedar oil (1.51) was about 63°.

It may be observed here that the tellurium precipitated from telluric acid by hydrazinehydrate is distinguished from that precipitated from a hydrochloric acid solution by sulphurdioxide which is also amorphous, by a perceptible darker colour. It is, as yet, undecided whether this is merely due to another degree of division or to a real allotropism of the amorphous modification.

§ 4. Both masses of tellurium mixed with 5—6 times the amount of powdered, freshly prepared potassium cyanide were fused for some hours in large Rose crucibles in an atmosphere of coal gas, with the aid of a large Perrot-furnace. In the course of a few months, about 5 kilos of these melts were obtained. When carefully powdered, the dark coloured masses dissolve in recently boiled, hot water to beautiful purple coloured solutions, which on cold oxidation by purified air deposit from the K_2Te all the tellurium in brilliant needles. On melting the masses, the poisonous influence of the hydrogen telluride, which is formed in small quantities, was experienced only too plainly, also the disagreeable consequences of breathing the very small quantities of $TeCl_2$ formed during the treatment with *aqua regia*. For weeks afterwards the breath has a powerful odour of $(CH_3)_2$ Te, which resembles phosphine and is exceedingly sensitive to the olfactory nerve of bystanders. [1])

The crystalline and already very pure tellurium thus obtained is free from selenium as proved by the exceedingly delicate Norris' potassiumiodide-reaction and by the non-reduction of the TeO_2 by hydroxylamine in strong hydrochloric acid solution. All the selenium has been removed as KCNSe, whilst the tellurium has passed into K_2Te and then has again been liberated by the action of air free from H_2S.

The purified element was now distilled in vacuo at about 600—700° in long tubes made of hard glass and containing plugs of asbestos: a Tégle furnace was used. This operation was repeated about seven times, each time about 10 grams were used. The pure tellurium thus obtained was silvery white and coarsely crystalline, much resembling crystallised antimony.

The determinations carried out have been made with the product obtained from telluric acid. The sulphur was recrystallised twice from boiling toluene and heated in a drying oven at 90° for some hours.

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[1]) The opinions as to the physiological actions of tellurium are still very much divided. Although selenium is an element hardly less poisonous than arsenic, tellurium is considered by Czapek and Weill (Chem N (1893), 1098 **2**) to be comparatively harmless, owing to the much more rapid reduction of the tellurium compounds and the consequent localisation in the organism. The experience gained in my laboratory proves this view to be incorrect.

Tellurium is undoubtedly poisonous, but the individual sensitiveness to small traces varies widely with different persons. TeH_2, in particular, is a poison causing severe headache and vomiting; other telluriumcompounds such as $TeCl_2$, for instance are supposed to cause much inconvenience only, owing to their conversion into malodorous substances, but still there can be no doubt whatever as to their poisonous nature.

§ 5. The construction of the melting apparatus will be readily seen from fig. 2. The hard glass tubes always filled with 10 grams of the weighed and well mixed complex of the two elements were placed in iron cylinders filled with fine sand. Tube and cylinder were covered with asbestos; the requisite atmosphere of nitrogen was supplied by way of a hard glass gas-inlet-tube. The nitrogen was prepared from NH_4Cl and KNO_2, freed from oxygen by means of alkaline pyrogallol and sodiumhydrosulphite and dried by sulphuric acid. The furnace was constructed of chamotte stone furnished

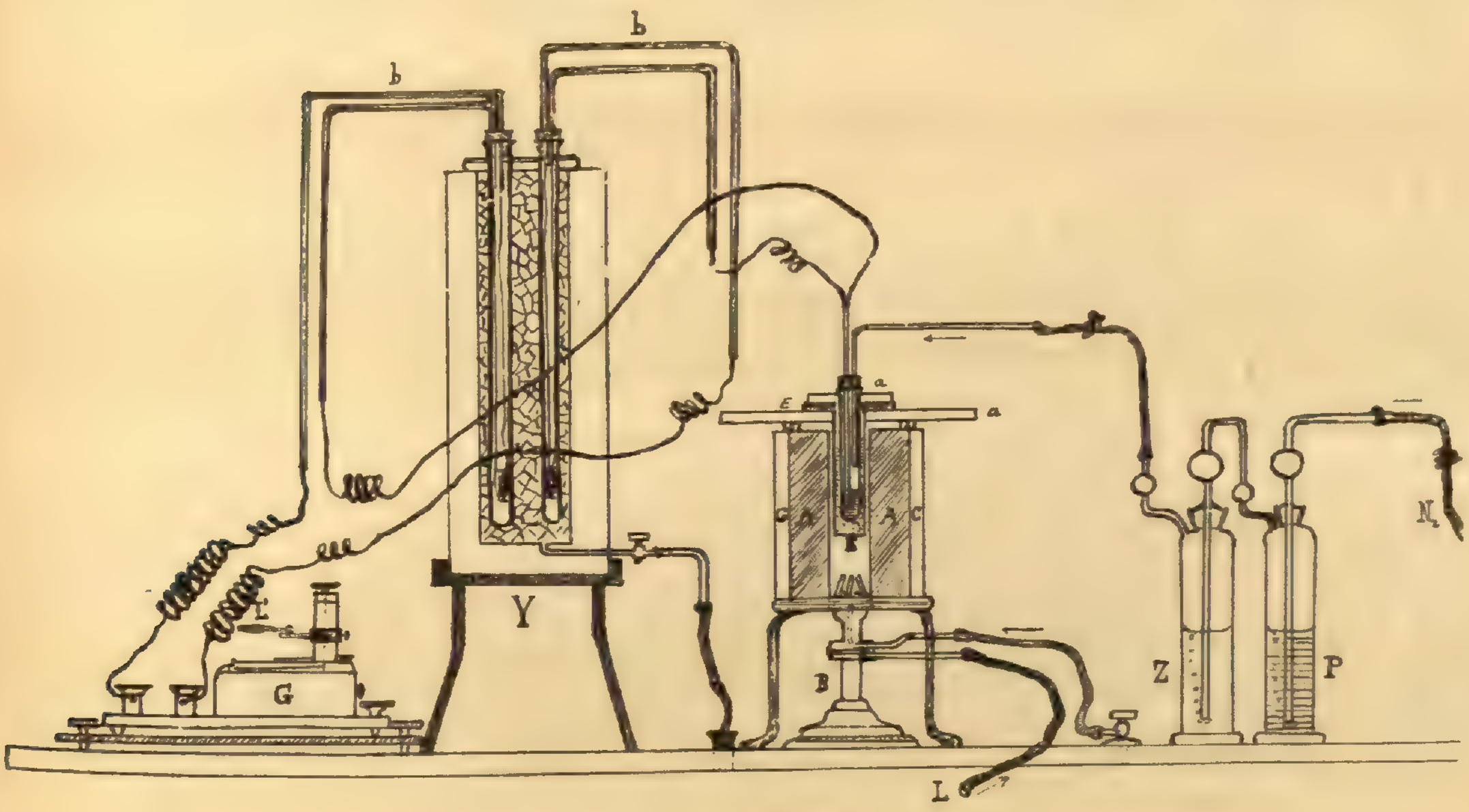

Fig 2.

with an asbestos filling and a central cylinder of unglazed earthenware; it was covered with an asbestos board resting on three little chamotte blocks, which were either removable or not so, for the regulation of the velocity of cooling. The icekettle for the cold solderplace of the platinum-platinumrhodium thermoelement (3 mm.) is double walled and allows of working for some six hours with the single supply of ice; all the conducting wires were isolated by glass tubes.

The galvanometer of SIEMENS and HALSKE was verified by determining the meltingpoints of tin, lead, bismuth, cadmium, zinc, antimony and silver and by making use of the values found by DAY and HOLBORN and by DAY and CLEMENT, which were compared with the gasthermometer. The reading was taken with the aid of a lens, the counting of the time by means of a clockwork, which gave a signal every 10 seconds.

§ 6. Great difficulties were experienced in the determination; when we dealt with mixtures containing much tellurium every precaution had to be taken to prevent the boiling off of the sulphur, and in the case of complexes containing much sulphur trouble arose from the great viscosity of the fusions and the very slow crystallisation of the masses. If the percentage of sulphur exceeds 80, the deter-

MELTING POINT DIAGRAM OF SULPHUR-TELLURIUM COMPLEXES.

Mol. % Sulphur	% by Weight Sulphur	Initial solidifying point C°	End solidifying point C°	Period of solidifying in seconds
0	0	452½	—	—
5	1.3	440	437	—
10	2.7	435	430	—
15	4.2	431	423	—
20	5.9	426	—	—
25	7.7	421	—	0
30	9.7	413	103	30
35	11.9	401	102	60
40	14.4	394	106	80
45	17.1	389	104	—
50	20	387	105	100
55	23.5	385	105	—
60	27.4	374	109	—
65	31.7	368	101	145
66.67	33.4	366	105 (106°)	160
70	36.9	361	105	—
75	43	348	108	180
80	50	339	109	180
85	58.7	—	110	210
90	69.3	288	108	230
95	82.7	—	109	260
98	92.4	—	110	—
100	100	115	—	—

minations often become very uncertain; some of these mixtures only exhibited a sharp end-solidifyingpoint. Still it was generally possible to get concordant results on repeating the experiments.

The subjoined table shows the results of the experiments.

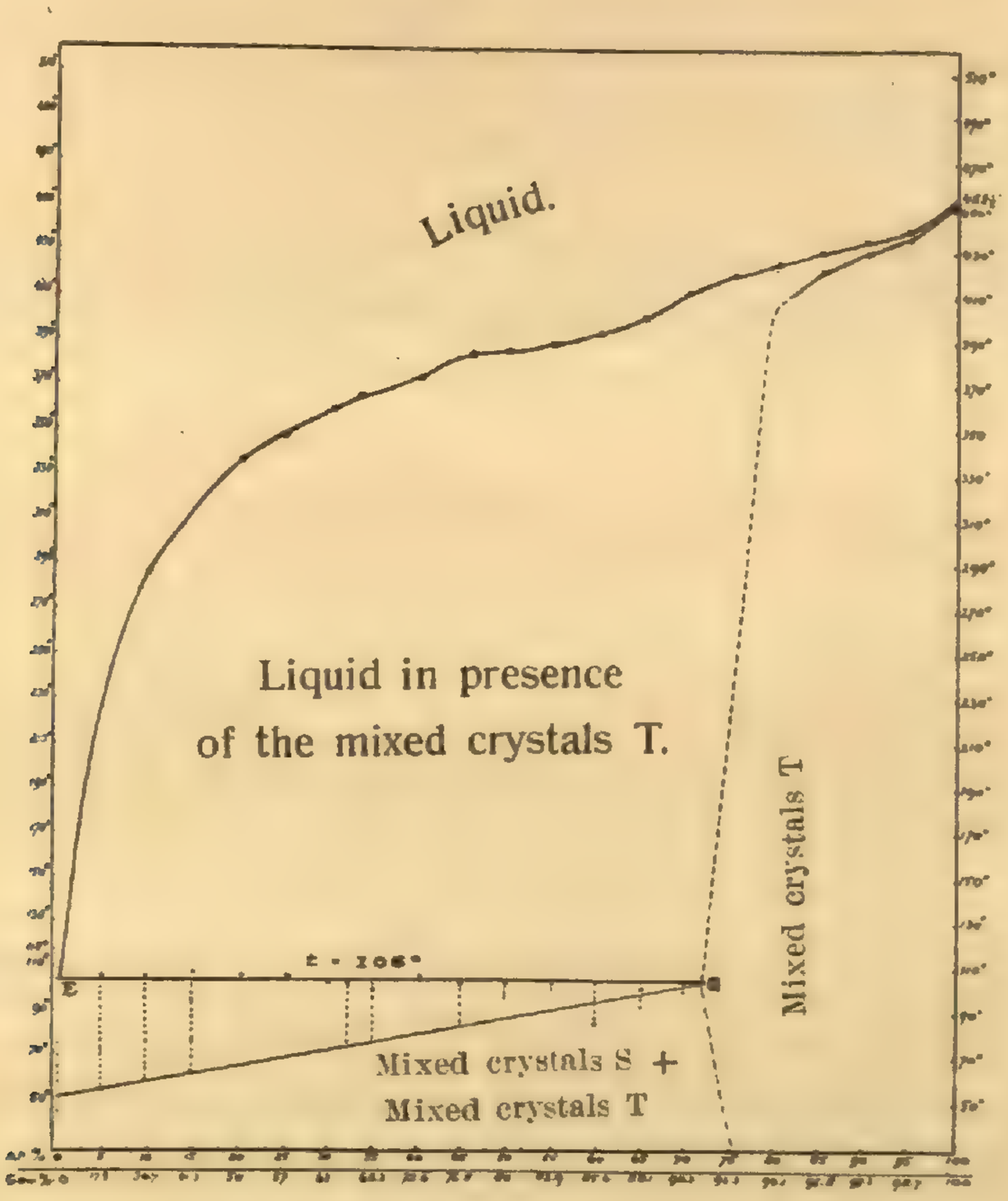

Fig. 3.

§ 7. These *data* represented graphically in Fig. 3 in the usual manner show, therefore, that the elements sulphur and tellurium when melted together yield, when solidifying, two series of mixed crystals of a different crystalline form. The condition diagram is that which has been noticed frequently with isodimorphous substances; there is a very extended hiatus starting from almost pure sulphur to perhaps 27% of sulphur at the side of the trigonal mixed crystals. The temperature of the eutectic point E is 106°; the time required for solidification, as far as could be ascertained, increases continuously with the percentage of sulphur until the pure sulphur is reached. The mixed crystals rich in sulphur have a slight ruddy colour; as very small amounts of tellurium impart to sulphur an intensely red colour, their tellurium content must be small indeed. They exhibit the thin needle shaped form of monoclinic sulphur. The transformation at 106° may be seen beautifully with the eye in the various melts on cooling as well as on warming. The monoclinic mixed crystals

rich in sulphur appear to change into the rhombic form at a lower temperature. In these circumstances nothing is noticed as to compounds between tellurium and sulphur; even at lower temperatures no heat effects are observed. The melts of the mixtures rich in tellurium are dark brownish black and in thin layers yellowish brown; unlike the melts rich in sulphur they are thin fluid up to their solidifying points.

§ 8. Considering all that is known up to the present as to the behaviour of the elements sulphur, selenium, and tellurium on being melted together, we may say that in this respect, tellurium certainly deserves the place assigned to it by DUMAS. Sulphur and selenium form, according to RINGER [1]), a trimorphous series of mixed crystals, selenium and tellurium, according to PELLINI and VIO [2]) an uninterrupted series of trigonal mixed crystals; but no compounds are formed, as may be expected, looking at the experience gained, apart from the exceptions in such triads of homologous elements, — at any rate in the central groups of the periodic system. With sulphur and selenium the matter is even somewhat still more complicated, as three instead of two heteromorphous kinds of mixed crystals occur in this case. If we accept RETGERS' view according to whom a less stable form, mostly unknown in the free state, of each of the components should correspond to each of these forms, the isotrimorphism in the case of selenium and sulphur is certainly more difficult to explain than the dimorphism of sulphur and tellurium. For of the two monoclinic series in the system: sulphur-selenium one, according to MUTHMANN, is analogous to the form of γ-sulphur, whereas the trigonal series would already possess the form of metallic selenium. But neither of the two known monoclinic modifications of selenium is isomorphous with any monoclinic modification of sulphur [3]), whilst the trigonal so-called ε-form of this element differs from the trigonal form of selenium. Looking from RETGERS' standpoint both these elements should be credited, in addition to their well known allotropic forms, with at least another two unknown, less stable modifications. In the trigonal series of the system: sulphur-tellurium we are dealing obviously with the same less stable trigonal form of sulphur as in RINGER's investigation, whilst the assumption of an unstable monoclinic form of tellurium cannot have anything artificial about it, in view of the fact that this symmetry occurs frequently

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[1]) RINGER, Z. f. anorg. Chem **32**. 183. (1902).

[2]) PELLINI and VIO, Gazz. Chim. It. (1906). **II**. 476.

[3]) GROTH, Chemische Krystallographie, Bd. I. (1906). p. 23—35.

both with selenium and sulphur. The research of PELLINI and VIO also does not introduce any further complications; the two elements are united there in all proportions to one trigonal series, so that only the sulphur-containing complexes of selenium and tellurium exhibit the hiati on mixing in the solid condition.

All this admits of the conclusion that the elements sulphur, selenium and tellurium form indeed a natural triad of perfectly homologous elements which are more adjacent to each other than their group-fellow *oxygen* is to any one of them. There can be only question of true "compounds" when one of the elements combines with oxygen [1]).

§ 9. Now there is still the question: what must be thought of the tellurium-sulphur complexes which are formed, at the temperature of the room, by means of H_2S from solutions of tellurites and tellurates, and in what sense must the so-called double sulphides obtained by OPPENHEIM and BERZELIUS be regarded.

First of all, I soon succeeded in showing that the element tellurium and particularly its amorphous modification dissolves, without leaving any residue, when heated with a solution of alkali- or ammonium-sulphide, also that the solubility increases with the concentration of the sulphide; and further that the solubility also increases when potassium hydroxide is added to the sulphide solution, thus retarding the hydrolysis. Clear yellow solutions are so formed turning somewhat ruddy on boiling, and oxidising rapidly in contact with the air with formation of a black precipitate. They are strongly alkaline and give with hydrochloric acid a heavy, black precipitate with evolution of H_2S; the precipitate appears to contain tellurium as well as sulphur and is soluble in alkalihydroxyde.

The analysis of these black precipitates did not give constant values; the tellurium content is much dependent on the *modus operandi* and oscillated between 46.9 % and 80.9 %.

Thereupon, the action of H_2S on different tellurium compounds was investigated: on the basic nitrate, on the finely divided dioxide suspended in absolute alcohol, on dioxide in hydrochloric acid solution, on telluric acid in water and on the tellurite- and tellurate-solutions obtained from TeO_2 or telluric acid. A beautiful, somewhat crystalline looking product was obtained from the alcoholic suspension of TeO_2; the analysis of the blue-black substance gave 80.1 %—80.9 % of tellurium whereas theory requires 79.9% for TeS, 66.6% for TeS_2

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[1]) It is, moreover, also known that O and S, for instance, never give isomorphous substitutions in organic compounds. S, Se and Te, however, behave differently as shown by the research of PELLINI, NORRIS, TUTTON, and others.

and 57% for TeS_3 so that the composition came very near to that of TeS, but with an *excess* of tellurium.

The telluric acid was not reduced in the cold; the TeO_2 dissolved in hydrochloric acid yielded a black precipitate with 71.2% of tellurium and therefore situated between TeS and TeS_2.

The basic nitrate is rapidly attacked by H_2S, but only at the surface; on the other hand it dissolved completely in boiling ammoniumsulphide, which solution, after being concentrated in vacuo at 40°, and allowed to crystallise in a vacuum desiccator over CaO, yields hard, pale yellow needles of a compound which may be recrystallised in vacuo without decomposition. The colour of the needles is greenish-yellow: they dissolve in water to a clear, yellow, strongly alkaline solution, which rapidly oxidises when exposed to the air; the crystals also soon turn black on exposure. The analysis gave a varying tellurium-content according to the method of preparation; in one instance were found 20.1% (NH_4), 42% Te and 37.9% S, which leads to the formula $(NH_4)_6Te_2S_7$ [1]).

In an analogous way the potassium compounds were prepared from the tellurite and tellurate with H_2S, by solution of the precipitate in the solution saturated with H_2S, or by solution in KOH, and by conduction of H_2S through it; the solutions were evaporated in vacuo, and were then left to crystallize over calcium oxide. Apparently the same yellow compounds are formed in all these cases, which crystallize in rosette-shaped aggregates of hard, fine needles, which in a humid condition smell strongly of H_2S, and yield clear, readily oxidisable solutions. Also the solid salts themselves oxidise rapidly, in which they become greenish yellow, and finally perfectly black. On close investigation the colour appears to assume different shades, even if to all appearances the same mode of preparation is used. Attempts to find a constant composition for these salts, have been unsuccessful; successively it was found to be: 35.4% Te (calculated

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[1]) The analysis of these complexes is a very tedious operation. If tellurium only has to be estimated and no sulphur, the reduction process with SO_2 or $NaHSO_3$ is still to be preferred. In our case, the tellurium had to be precipitated from a boiling, strongly ammoniacal solution with hydrazine hydrate, which reduction proceeds very slowly and also incompletely, and had often to be repeated half a dozen times. The last traces of still dissolved tellurium betray themselves on heating by the fine steel blue colour of the colloidal tellurium present; this is generally completely precipitated on rendering the liquid acid, and by way of control the solution may then be heated once more with ammonia and hydrazine hydrate. The tellurium was collected on a weighed filter dried at 100° and weighed as such. The SO_3 was weighed as $BaSO_4$, the K as KCl or $KClO_4$ the NH_3 as NH_4Cl. For obvious reasons the analysis of the barium salt is a very tedious affair.

for $K_6Te_2S_7$ 35.7%); 33.5% Te, 33.4% S and 33.1% K, which answers to a formula $K_{10}Te_3S_{12}$; another time (for a product prepared from K_2TeO_4): 44.7% Te, 31.47% S, and 23.7% K, which would correspond to $K_{12}S_{20}Te_7$; again another time at somewhat higher temperature: 37.5% Te, 34.3% S and 28.1% K, which leads to a formula $K_{12}Te_5S_{19}$.

The behaviour is practically analogous to that found in the polysulphides of the alkalies towards sulphur, where, according to KÜSTER's researches [1]), very complicated equilibria between different polysulphides and their dissociation products occur in the solution; and to that of selenium towards sulphides where, according to MESSINGER [2]), a portion of the sulphur of the complex sulphohydrogen sulphides may be replaced by selenium, forming such compounds as Na_2SSe_2 which, therefore, belong to the type of a *tri*sulphide. The behaviour of amorphous tellurium towards sulphide solutions as described previously also agrees with the notion that we are dealing here with salts of complex tellurohydrogen sulphides which in such solutions are in dissociation-equilibria with each other and are moreover split hydrolytically.

The type of the *tri*sulphides becomes then of particular importance next to that of the *di*sulphides: $K_{10}S_{12}Te_3$ may be derived from K_2S_3 by isomorphous substitution of $^1/_5$ of the S by Te; $K_6Te_2S_7$ and $(NH_4)_6Te_2S_7$ similarly from K_2S_3, or $(NH_4)_2S_3$; on the other hand $K_{12}Te_5S_{19}$ has again the character of the type K_2S_4 etc.

§ 10. Although these compounds do not as a rule occur in measurable forms (the K-salt was obtained a few times as beautiful rhombic parallelopipeds with faint double refraction and without perceptible dichroism) I finally succeeded in obtaining very large, yellow crystals, with many planes, of a barium salt prepared by dissolving the black precipitate formed by the action of H_2S on potassium tellurite in BaS-solution. The analysis indeed, did not always yield precisely the same results, but still the composition agreed closely with the formula $Ba_3S_7Te_2$; in one instance the normal composition; 45,8% Ba; 25% S and 29,1% Te was actually found. These crystals were accurately investigated and proved to be so well constructed that, in their habit, they did not remind us of mixed crystals but, on the contrary, made a vivid impression of belonging to a true chemical compound. The following data were found:

Large yellow transparent crystals fairly stable in the air but, after some time, assuming a greyish colour. They are well constructed,

[1]) KÜSTER, Z. f. anorg. Chem. **44**. 431.
[2]) MESSINGER, Berl. Ber. **30** 805 (1897).

yield constant angular values and have, crystallonomically, quite the appearance of a well defined compound of constant composition.

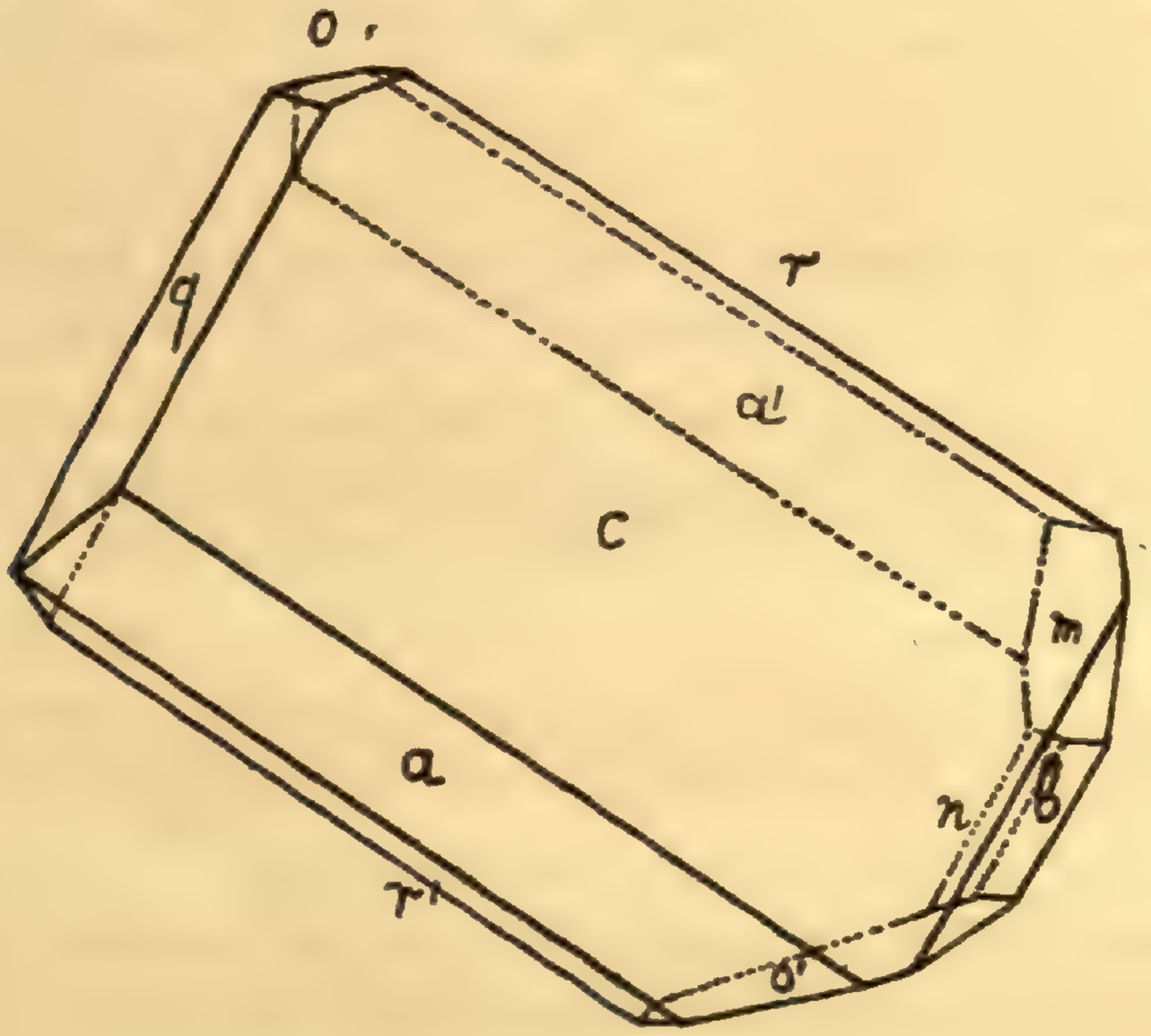

Fig. 4.

The compound has, in moist air, a strong odour of hydrogen-sulphide and is decomposed by water with separation of a black substance which contains tellurium and sulphur.

Triclinic-pinacoïdal.

$$a : b\ c = 1.6835 : 1 : 1.5515$$

$A = 109° 43'$	$\alpha = 113° 7\frac{1}{2}'$
$B = 122° 10\frac{1}{2}$	$\beta = 124° 13'$
$C = 90° 32'$	$\gamma = 77° 39'$

Forms observed: $c = \{001\}$, strongly predominating; $a = \{100\}$ and $b = \{010\}$ equally well developed and lustrous; $q = \{0\bar{1}1\}$ and $r = \{\bar{1}01\}$ quite as much developed as a and b, and yielding sharp reflexes; $o = \{11\bar{2}\}$, lustrous and fairly large; $n = \{01\bar{2}\}$ small but lustrous; $\omega = \{\bar{1}11\}$, small and subordinate, generally with but one plane; $m = \{\bar{1}10\}$, well developed and lustrous, also without the parallel opposite plane.

The habit is flattened towards {001} with slight stretching in the direction of the b-axis. A complete or distinct plane of cleavage is not found.

The following angular values were measured:

	Measured :	Calculated :
$a : b = (100) : (010) =$	*89° 28′	—
$b : c = (010) : (001) =$	*70 17	-
$c : a = (001) : (100) =$	*57 49½	—
$c : r = (001) : (\bar{1}01) =$	*53 46½	—
$c : q = (001) : (0\bar{1}1) =$	*65 45	—
$o : b = (11\bar{2}) : (010) =$	67 32	67° 36′
$r : b = (\bar{1}01) : (010) =$	68 47	68 47½
$a : q = (100) : (0\bar{1}1) =$	67 29	67 26
$c : o = (001) : (\bar{1}\bar{1}2) =$	50 5	49 49
$a : r = (\bar{1}00) : (\bar{1}01) =$	68 24	68 24
$c : n = (00\bar{1}) : (01\bar{2}) =$	38 17	38 27
$n : q = (01\bar{2}) : (01\bar{1}) =$	27 24	27 18
$q : b = (01\bar{1}) : (010) =$	43 56	43 58
$r : o = (10\bar{1}) : (11\bar{2}) =$	52 35	52 47
$o : q = (11\bar{2}) : (01\bar{1}) =$	32 21	32 19
$m : b = (\bar{1}10) : (010) =$	33 51	33 37
$m : a = (\bar{1}10) : (\bar{1}00) =$	56 41	56 55
$m : r = (\bar{1}10) : (\bar{1}01) =$	59 37	59 32½
$m : q = (\bar{1}10) : (01\bar{1}) =$	35 10	35 21½
$\omega : b = (1\bar{1}1) : (010) =$	40 55	40 47
$\omega : c = (11\bar{1}) : (00\bar{1}) =$	85 46½	85 48½
$\omega : a = (11\bar{1}) : (100) =$	74 28	74 30½
$\omega : q = (11\bar{1}) : (01\bar{1}) =$	38 5	38 3½

The agreement between the observation and the calculation is an excellent one.

Etching figures were not obtained. It may be, — looking at the acentric habit and plane-development of some of the forms, — that the symmetry is triclino-*pedial*. The situation of the optical axial angle could not be determined. That of the optical main directions was such that the angle of extinction on {001} with the side (001) : ($\bar{1}$01), was about 15°, but on {$\bar{1}$01} with the same side it amounted to about 12°, and that with an inclination which on {101} proceeds from the left in front to the right at the back, and on {001} from the right above to the left below.

Here we are, consequently, also dealing with a polysulphide of the type BaS_2, in which ²/₃ of the sulphur has been replaced by tellurium.

Efforts to obtain this compound, prepared from Ba S and S, in a measurable form, and thus to obtain an argument in favour of the said view, in the event of an isomorphism between the two substances, have been found so far unsuccessful.

§ 11. In the electrolysis of a dilute solution of the potassium salt, in which the platinum-dish acted as the cathode and a disc-shaped platinum-electrode as anode, it looked as if tellurium was precipitated at both electrodes. The liberation of the black substance at the anode is nothing else than an oxidationphenomenon.

The tension at the electrodes was 2.6 Volt, the current 0.05 Ampère; the oxygen formed at the anode oxidises the liquid, so sensitive to oxygen, with separation of tellurium-sulphur complexes which partly stick to the anode and partly collect above the same on the liquid; if the current passes for some time the precipitate redissolves and the anode again turns bright. I have ascertained that the adhering precipitate contains tellurium as well as sulphur.

On the other hand, the precipitation of a black substance at the cathode takes place continuously but very slowly; after twelve hours only a small portion of the salt, about one gram and a half in 50 cc of distilled water, had been decomposed by the current. This black precipitate has now proved to be pure tellurium and this observation would, therefore, go against the assumption that tellurium forms a constituent of the anion. This experiment, however, *cannot* be used as evidence against that view, since we know an analogous case in the electrolysis of sodium-sulphantimonate [1]), where the antimony also proceeds, apparently, to the cathode, although it acts, in the salt, as a constituent of the anion.

It has also transpired in these experiments that the metal does not wholly take part in the electric conductivity, but that in the electrolysis of the solution, the sodium sulphide is decomposed, and either the sodium liberated at the cathode, or the hydrogen which it causes to be evolved, precipitates the antimony by a secondary reaction. Only when a very little alkalisulphide is present, the anion $''SbS_4$ also moves towards the anode. Obviously, the explanation in our case is the same; the tellurium is formed secondarily at the cathode, whilst at the anode, as in the experiments cited, fairly complicated and somewhat obscure oxidationphenomena occur. In each case, this apparent contradiction does by no means prove the incorrectness of the view, that the said salts may be considered as derivatives of

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1) Ost und Klapproth, Zeitschr. f. angew. Chemie (1900). p. 827.

complex tellurohydrogen sulphides. The investigation of this exceedingly complicated subject is being continued in the direction indicated.

§ 12. Summarizing the results of these investigations, I believe I may say:

1. That the elements tellurium and sulphur do not form compounds, but mixed crystals.

2. That the elements tellurium, selenium and sulphur behave in quite an analogous manner towards the sulphides of the alkali and alkali-earth metals, and cause the formation of complex sulpho-, seleno- or tellurohydrogen sulphides of a different type, and that it is quite unnecessary to presuppose the intermediate formation of selenium-sulphur or tellurium-sulphur compounds.

3. That the position, given by DUMAS, to tellurium in the sulphur group as the first homologue of selenium is quite justified so far as the mutual behaviour of these elements is concerned, and that sulphur, selenium, and tellurium form a natural triad of elements, which are more adjacent to each other than any one of them is to oxygen.

Groningen, Inorg. Chem. Lab. of the University.

Physics. — "*Some remarks on Prof.* KOHNSTAMM's *reply.*" By Dr. J. J. VAN LAAR. Communicated by Prof. LORENTZ.

In these Proceedings of Jan. 6th 1910 Prof. KOHNSTAMM has inserted a reply to my remarks suggested by a paper by Messrs. TIMMERMANS and KOHNSTAMM. Though I, too, very reluctantly continue the discussion, I feel obliged to briefly revert to this matter for the last time, in order to prevent further misunderstanding.

So I will just point out that Mr. KOHNSTAMM is quite silent about the *cardinal* point of my remarks, given in point *a* second part, point *e* and point *f*; viz. that Messrs. T. and K. in consequence of their disregard of the last five of my seven papers on the subject in question have wrongly asserted that the "abnormal" type III could not occur for *normal* substances, and that this would be due to my restricting supposition $a_{12} = \sqrt{a_1 a_2}$. Only to remove this misunderstanding — as I had asserted the very opposite of this — I wrote my preceding paper.

On the other hand a few minor questions are extensively discussed in the answer, viz. the question $a_{12} = \sqrt{a_1 a_2}$ and $\frac{d^2b}{dx^2} = 0$. I must remark here that when I repeatedly spoke of the "quite general" case $a_2 \gtrless a_1$, $b_2 \gtrless b_1$, this expression "quite general" was obviously meant in *contrast* to the special case $a_2 \gtrless a_1$, $\underline{b_2 = b_1}$, treated by me before in the first two Papers, as would be clear to everybody, and that the "generality" meant by me according *to the whole tenour* of my papers, of course, only holds *within the area of the once assumed supposition* $a_{12} = \sqrt{a_1 a_2}$ (BERTHELOT's). For this supposition I explicitly premised in all my papers, and I repeated it more than once (loc. cit.).

Now with regard to the question itself of the supposition $a_{12} = \sqrt{a_1 a_2}$ (which, however, is not the point at issue), I may be allowed to remind Mr. K. of a paper of his in the Zeitschr. f. physik. Ch. **36** p. 41 (1901), where it, inter alia, says at the end (p. 62): "So weit ich aus dem mir bekannten Material zu schlieszen vermag, scheinen mir jedoch die Tatsachen sehr zu Gunsten der (BERTHELOT'sche) Annahme zu sprechen ...." I will add that I, too, consider the supposition $a_{12} = \sqrt{a_1 a_2}$ as very probable, and that seeming deviations from this supposition are attributed by me to the formation of compounds. But I hope to treat this more fully on a later occasion.

I now demonstrated that even on this simple supposition the abnormal type III can occur for *perfectly normal* substances. And this Mr. K. denied — as my later papers on this subject in which this was proved by me, had evidently escaped his notice.

With regard to the supposition $\frac{d^2b}{dx^2} = 0$, Mr. K. refers to my statement that "qualitatively everything will remain the same if b is not assumed independent of v and T". This, however, is quite beside the question whether the supposition $\frac{d^2b}{dx^2} = 0$ is of influence on my results or not; for v and T are not the same thing as x. I fully maintain my contention, and Mr. K. will, no doubt, understand, that this dependence on v and T was only mentioned by me, because VAN DER WAALS' later investigations have shown that b still depends on this quantity. But this is not the point in question.

I, however, readily acknowledge that when writing the lines about the longitudinal plait closing again, quoted by Mr. K., I did

not sufficiently clearly state that the divergent result was only founded on the assumption $\frac{d^2b}{dx^2} = 0$. I knew, however, that VAN DER WAALS in his Continuität II p. 24 has already treated this question. Yet on theoretical considerations I abide by my opinion that in the neighbourhood of the limiting volume, so at very high pressures, $\frac{d^2b}{dx^2}$ must be $= 0$.

And now I think that I for my part, have sufficiently elucidated Mr. KOHNSTAMM's Reply, so that further misunderstanding seems almost precluded.

Baarn, Febr. 21, 1910.

Mathematics. — *"The oscillations about a position of equilibrium where a simple linear relation exists between the frequencies of the principal vibrations."* (1st part). By Mr. H. J. E. BETH. (Communicated by Prof. KORTEWEG).

Introduction.

§ 1. In his paper[1]): "On certain vibrations of higher order of abnormal intensity (vibrations of relation) for mechanisms with more degrees of freedom" (Verhandelingen der Koninklijke Akademie van Wetenschappen, Vol. V. N°. 8, 1897; Archives Néerlandaises Vol. I, series II, pages 229—260) Prof. KORTEWEG has written down the expansions in series for the principal coordinates of an arbitrary mechanism with more degrees of freedom, performing small oscillations about a position of stable equilibrium. From these expansions in series could be deduced that in a certain case it was possible for some vibrations of higher order, having in general a small intensity with respect to the principal vibrations, to obtain an abnormally great intensity; this is the case when between the frequencies n , n_y etc. of the principal vibrations a relation exists of the form

$$pn_x + qn_y + \ldots = \varrho;$$

where p, q etc. are positive or negative integers and ϱ is with respect to n_x, n_y etc. a small quantity, called residue of relation.

Furthermore however it became evident that, when $S \leqq 4$ (S is the sum of the absolute values of p, q etc.) and at the same time $\varrho = 0$,

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[1]) "Over zekere trillingen van hooger orde van abnormale intensiteit (relatietrillingen) bij mechanismen met meerdere graden van vrijheid".

the above-mentioned expansions in series lost their validity; we must therefore investigate in a different way what becomes of the movement in the case mentioned. In what follows we shall investigate this for a mechanism with two degrees of freedom. As a base for this investigation a very simple mechanism is selected, namely a material point which moves without friction yet under the influence of gravitation on a given surface in the vicinity of its lowest point. Every time one of the cases $S \leqq 4$ is discussed we shall pass to an arbitrary mechanism with two degrees of freedom.

Movement on the bottom of a surface.

§ 2. We shall accordingly first pass on to the treatment of the simple mechanism we have chosen as a base for our investigation. When the surface has positive curvature in the vicinity of its lowest point O, when plane XY is the tangential plane in O, and the XZ- and YZ-planes are the principal sections of the surface in that point, whilst the Z-axis is supposed positive upwards, then the equation of the surface in the vicinity of O takes the form of:

$$z = \frac{1}{g}(c_1 x^2 + c_2 y^2 + d_1 x^3 + d_2 x^2 y + d_3 x y^2 + d_4 y^3 + \ldots); \quad . \quad (1)$$

where c_1 and c_2 are positive.

The equations of motion of the material point become:

$$\left.\begin{aligned} \ddot{x} + \frac{\partial z}{\partial x}(g + \ddot{z}) &= 0, \\ \ddot{y} + \frac{\partial z}{\partial y}(g + \ddot{z}) &= 0. \end{aligned}\right\}$$

Availing ourselves of (1) to eliminate $\ddot{z}$ we find:

$$\left.\begin{aligned} \ddot{x} + \frac{\partial z}{\partial x}(g + \frac{\partial^2 z}{\partial x^2}\dot{x}^2 + 2\frac{\partial^2 z}{\partial x \partial y}\dot{x}\dot{y} + \frac{\partial^2 z}{\partial y^2}\dot{y}^2 + \frac{\partial z}{\partial x}\ddot{x} + \frac{\partial z}{\partial y}\ddot{y}) &= 0, \\ \ddot{y} + \frac{\partial z}{\partial y}(g + \frac{\partial^2 z}{\partial x^2}\dot{x}^2 + 2\frac{\partial^2 z}{\partial x \partial y}\dot{x}\dot{y} + \frac{\partial^2 z}{\partial y^2}\dot{y}^2 + \frac{\partial z}{\partial x}\ddot{x} + \frac{\partial z}{\partial y}\ddot{y}) &= 0. \end{aligned}\right\} \quad (2)$$

Let h be the small quantity (small e.g. with respect to the principal radii of curvature R_1 and R_2 of the surface in O) which determines the order of greatness of x and y, then the equations (2) become, omitting the terms of order h^2 and higher:

$$\left.\begin{aligned} \ddot{x} + 2c_1 x &= 0, \\ \ddot{y} + 2c_2 y &= 0. \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

These equations are in general sufficient to arrive at the solution at first approximation. This then becomes:

$$\left.\begin{aligned} x &= Ah\cos(n_1 t + \lambda), \\ y &= Bh\cos(n_2 t + \mu); \end{aligned}\right\} \quad \ldots\ldots\ldots \quad (4)$$

where $n_1 = \sqrt{2c_1}$, $n_2 = \sqrt{2c_2}$.

Here Ah, Bh, λ and μ are constants of integration; we suppose A and B to be of moderate greatness.

At first approximation therefore the horizontal projection of the moving point describes a Lissajous curve, which is closed when $pn_1 = qn_2$, where p and q are integers. If $pn_1 = qn_2 + \varrho$, the curve described is not closed, but it consists of a succession of parts each of which differs but little from a closed curve. These last closed curves have however various shapes which answer to different values of the difference in phase. They are all described in the rectangle with $2Ah$ and $2Bh$ as sides.

§ 3. If we wish to take into consideration the terms of a higher order appearing in (2) we generally have but to apply small modifications to the first approximation.

These modifications are, however, not small in case a relation exists of the form:

$$pn_1 = qn_2 + \varrho;$$

where $S \equiv p + q \leqq 4$ and $\frac{\varrho}{n_1}$ is very small (what is meant here by "very small" will be evident later on).

When by applying the method of consecutive approximations, starting from (4) as first approximation, we try to find expansions in series for x and y, we shall find, when substituting the expressions (4) into the terms of higher order of (2) and developing the products and powers of the cosines, in case $\frac{\varrho}{n_1}$ is very small, periodical terms which have about the same period as the principal vibration, to which the equation in which the indicated term appears relates more especially. Such terms in the equations of motion give rise in the expansions in series for x and y to terms with abnormally great amplitude. These amplitudes may reach the order h and even seem to be greater still.

This proves that in the case supposed our first approximation was not correct. It is evident that in the equations of motion there are terms of higher order, which are of influence even on the first

approximation. So we shall have to find in the equations (2) which terms give rise to the failure of the application of the method of consecutive approximations. These terms we shall have to include in the abridged equations, serving to determine the first approximation.

We shall consecutively discuss the cases:

$$S = 3\ (2n_1 = n_2 + \varrho), \qquad S = 4\ (3n_1 = n_2 + \varrho), \qquad S = 2\ (n_1 = n_2 + \varrho).$$

$S = 3$.[1]) *Strict relation.*

§ 4. We suppose $\varrho = 0$; therefore

$$n_2 = 2n_1.$$

In the equations of motion appear for the first time among the terms of order h^2 terms which, according to what was said in § 3, must be included in the abridged equations. They are: in the first equation $2d_2 xy$, in the second $d_2 x^2$. These are the most important among the terms referred to. Omitting the remaining terms of higher order we therefore have to consider:

$$\left.\begin{aligned} \ddot{x} + n_1^2\, x + 2d_2\, xy &= 0, \\ \ddot{y} + 4n_1^2\, y + d_2\, x^2 &= 0. \end{aligned}\right\} \quad \ldots\ldots \quad (5)$$

We may also write this system as follows:

$$\left.\begin{aligned} \ddot{x} + n_1^2\, x - \frac{\partial R}{\partial x} &= 0, \\ \ddot{y} + 4n_1^2\, y - \frac{\partial R}{\partial y} &= 0; \end{aligned}\right|$$

in which:

$$R \equiv - d_2\, x^2\, y.$$

To this we apply the method of the variation of the canonical constants. This means, as is known, that the equations, arising when the terms $\frac{\partial R}{\partial x}$ and $\frac{\partial R}{\partial y}$ are omitted, first are solved; in which solution 4 arbitrary constants appear; we then investigate what functions of the time must be the quantities just now regarded as constants, so that the expressions for x and y, taken in this way, represent the solution of the complete equations containing $\frac{\partial R}{\partial x}$ and $\frac{\partial R}{\partial y}$. The equations in which $\frac{\partial R}{\partial x}$ and $\frac{\partial R}{\partial y}$ are lacking, are solved according to

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[1]) In a following paper we shall discuss the cases $S = 2$ and $S = 4$.

the method of HAMILTON-JACOBI in order that the constants we obtain may form a canonical system.

If α_1, α_2, β_1 and β_2 are the canonical constants then by substitution of the expressions found for x and y in R this R will become a function of α_1, α_2, β_1, β_2 and t. The variability of the α's and β's with the time is then given by:

$$\frac{d\alpha_1}{dt} = \frac{\partial R}{\partial \beta_1}\ ,\quad \frac{d\alpha_2}{dt} = \frac{\partial R}{\partial \beta_2}\ ,\quad \frac{d\beta_1}{dt} = -\frac{\partial R}{\partial \alpha_1}\ ,\quad \frac{d\beta_2}{dt} = -\frac{\partial R}{\partial \alpha_2} \quad . \quad (6)$$

In case R is a function of the α's and the β's alone, and consequently does not contain t explicitly, the system has as an integral:

$$R = \textit{constant}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

§ 5. If now we solve the equations

$$\left.\begin{aligned} \ddot{x} + n_1^2\, x &= 0, \\ \ddot{y} + 4n_1^2\, y &= 0, \end{aligned}\right\}$$

arising from (5) by omission of the terms $\frac{\partial R}{\partial x}$ and $\frac{\partial R}{\partial y}$, according to the method HAMILTON-JACOBI we may arrive at:

$$\left.\begin{aligned} x &= \frac{\sqrt{\alpha_1}}{n_1}\cos(n_1 t + 2n_1\beta_1), \\ y &= \frac{\sqrt{\alpha_2}}{2n_1}\cos(2n_1 t + 4n_1\beta_2); \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

where α_1, α_2, β_1, β_2 form a canonical system of constants. We must suppose α_1 and α_2 to be of order h^2 as the amplitudes of the x- and y-vibrations must be of order h.

Substitution of (8) in $R \equiv -d_2 x^2 y$ furnishes 3 terms:

$$\frac{\alpha_1\sqrt{\alpha_2}}{4n_1^3}\cos(2n_1 t + 4n_1\beta_2),\ \frac{\alpha_1\sqrt{\alpha_2}}{8n_1^3}\cos\{4n_1 t + 4n_1(\beta_1+\beta_2)\}\ \text{and}$$

$$\frac{\alpha_1\sqrt{\alpha_2}}{8n_1^3}\cos 4n_1(\beta_1-\beta_2),$$

each term multiplied by $-d_2$.

The first two terms contain t explicitly; setting aside the variability of the α's and β's we can say that those terms are periodical, whilst the period is comparable to that of the principal vibrations. The last term, however, does not contain t explicitly. Only this last term is of importance for the first approximation; the two others we omit (we shall revert to this in § 6).

We therefore take:

$$R = -\frac{d_2}{8n_1^2}\,\alpha_1 \sqrt{\alpha_2}\,\cos 4n_1(\beta_1-\beta_2).$$

Consequently system (6) takes this form:

$$\left.\begin{aligned}
\frac{d\alpha_1}{dt} &= \quad 2Nm_1\,\alpha_1\,\alpha_2^{\frac{1}{2}}\sin\varphi,\\
\frac{d\alpha_2}{dt} &= -2Nm_1\,\alpha_1\,\alpha_2^{\frac{1}{2}}\sin\varphi,\\
\frac{d\beta_1}{dt} &= \quad m_1\,\alpha_2^{\frac{1}{2}}\cos\varphi,\\
\frac{d\beta_2}{dt} &= \tfrac{1}{2}m_1\,\alpha_1\,\alpha_2^{-\frac{1}{2}}\cos\varphi;
\end{aligned}\right\} \quad \ldots\ldots (9)$$

where N is written for $n_2 = 2n_1$; further:

$$m_1 = \frac{d_2}{N^2},$$

$$\varphi = 2N(\beta_1-\beta_2).$$

As t does not appear explicitly in R we get according to what has been said at the close of § 4 as an integral:

$$\alpha_1 \sqrt{\alpha_2}\cos\varphi = constant. \quad \ldots\ldots (10)$$

Furthermore it appears at once from (9) that:

$$\frac{d\alpha_1}{dt} + \frac{d\alpha_2}{dt} = 0.$$

Therefore:

$$\alpha_1 + \alpha_2 = constant \quad \ldots\ldots (11)$$

is another integral.

The latter gives us reason to introduce a new variable ζ, in such a way that:

$$\alpha_1 = \frac{1}{4}R_0^2N^2h^2\zeta \quad , \quad \alpha_2 = \frac{1}{4}R_0^2N^2h^2(1-\zeta);$$

ζ is then always situated between 0 and 1, R_0 is of moderate greatness.

By this (10) obtains the form:

$$\zeta\sqrt{1-\zeta}\cos\varphi = K; \quad \ldots\ldots (12)$$

in which K represents a constant.

The first equation of (9) becomes:

$$\frac{d\zeta}{dt} = \frac{d_2R_0}{N}\zeta\sqrt{1-\zeta}\sin\varphi\,.\,h. \quad \ldots\ldots (13)$$

By elimination of φ between (12) and (13) we arrive at:

$$\frac{d\zeta}{\sqrt{\zeta^2(1-\zeta)-K^2}} = \pm\frac{d_2R_0}{N}h\,.\,dt.$$

Now put:

$$f(\zeta) \equiv \zeta^2 (1-\zeta) - K^2,$$

then for the initial value of ζ we find $f(\zeta) > 0$. For $\zeta = 0$ and $\zeta = 1$ we find $f(\zeta) < 0$. Thus the equation $f(\zeta) = 0$ has two roots between 0 and 1.

So K^2 cannot have all values; the possible values of K^2 lie between two limits; in § 9 we shall revert to this and to the special cases, corresponding to the limiting values of K^2.

The roots between 0 and 1 which the equation

$$\zeta^2 (1-\zeta) - K^2 = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (14)$$

has in the general case will be called ζ_1 and ζ_2, where we suppose $\zeta_2 > \zeta_1$. The third root is negative, we call it $-\lambda$.

The differential relation between ζ and t may now be written:

$$\frac{d\zeta}{\sqrt{(\zeta_2-\zeta)(\zeta-\zeta_1)(\zeta+\lambda)}} = \pm \frac{d_2 R_0}{N} h \, . \, dt. \quad . \quad . \quad . \quad . \quad (15)$$

So with the aid of elliptic functions ζ may be expressed in t. It changes periodically between the limits ζ_1 and ζ_2.

Now with the aid of (12) we can also calculate φ as function of t. And β_1 and β_2 likewise, it being possible to write the last two equations of (9):

$$\frac{d\beta_1}{dt} = \frac{d_2 R_0 K}{2N^2} \cdot \frac{h}{\zeta},$$

$$\frac{d\beta_2}{dt} = \frac{d_2 R_0 K}{4N^2} \cdot \frac{h}{1-\zeta}.$$

So now x and y are also known as functions of t [1]).

In fig. 1 the relation (12) between ζ and φ is represented in polar coordinates, φ is taken as polar angle, $\sqrt{1-\zeta}$ as radius vector. The circle drawn has unity as radius. The curves change with the value of K. For $K > 0$ the curves lie to the right of the straight line $\varphi = \frac{\pi}{2}$, for $K < 0$ to the left of it; $K = 0$ furnishes degeneration into the straight line $\varphi = \frac{\pi}{2}$ and the circle $\zeta = 0$. By the maximal positive and negative value of $K (K = \pm \frac{2}{9}\sqrt{3})$ the curve has contracted into an isolated point. The special cases of the motion belonging to $K = 0$ and to $K = \pm \frac{2}{9}\sqrt{3}$ will be discussed in § 9.

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[1]) These calculations will be found in my dissertation, which will appear before long.

§ 6. When astronomers try to obtain in the Theory of the disturbances of the movements of the planets by the application of the method of LAGRANGE expansions in series for the coordinates of the planets or the elements of their orbits, then terms may appear with abnormally large coefficients in consequence of small divisors, originating from the integration. This takes place when between the inverse values of the periods of revolution of some planets a linear relation with integer coefficients is almost fulfilled. Besides some other properties the terms are also distinguished according to their class, by which is meant:

$$\alpha - \frac{m}{2} - \frac{m'}{2};$$

where α represents the exponent of μ (a small quantity indicating the order of greatness of the disturbing function), m the exponent of t, m' the exponent of the small divisor, as they appear in the coefficient of the term indicated. Now it is the terms of the lowest class which we have to take into consideration if we wish to make the expansions in series to hold for a long space of time. By DELAUNAY a method is indicated to determine the terms of the lowest class. It consists principally in omitting all terms of short period (period comparable to the periods of the revolution of the planets) in the disturbing function and retaining the most important of the others. (Comp. e. g. H. POINCARÉ, Leçons de mécanique céleste, vol. I, page 341).

The problem under discussion has much resemblance with the one mentioned from the theory of disturbances. In the preceding § in omitting some terms in R we have imitated what is done in the theory of disturbances.

It is easy to see that the terms omitted have really no influence on the first approximation, when we consider the terms which appear e. g. in α_1 by introduction of such a term.

Osculating curves.

§ 7. In § 5 we have found that the movement of the horizontal projection of the material point might be represented by:

$$x = \frac{\sqrt{\alpha_1}}{n_1} \cos(n_1 t + 2n_1\beta_1),$$

$$y = \frac{\sqrt{\alpha_2}}{2n_1} \cos(2n_1 t + 4n_1\beta_2);$$

where α_1, α_2, β_1 and β_2 are slowly variable; for $\frac{d\alpha_1}{dt}$ and $\frac{d\alpha_2}{dt}$ are of order h^3, $\frac{d\beta_1}{dt}$ and $\frac{d\beta_2}{dt}$ of order h. (Comp. (9)).

For every arbitrary moment the α's and the β's have a definite value. These values determine a certain Lissajous curve. This curve we shall call the osculating curve for the moment indicated, which name is in use in the theory of disturbances. (See among others H. POINCARÉ, Leçons de mécanique céleste, vol. I, page 90). Thus in our problem the osculating curves are the wellknown Lissajous figures for 2 octaves.

By the change of the origin of time we may write the equations of an osculating curve:

$$x = R_0 h \sqrt{\zeta} \cos n_1 t,$$

$$y = \tfrac{1}{2} R_0 h \sqrt{1-\zeta} \cos (2n_1 t - \varphi);$$

where as in § 5 we have introduced ζ instead of α_1 and α_2; here too φ means $4n_1(\beta_1 - \beta_2)$.

We now see that φ is the value of the difference in phase, to which the osculating curve corresponds when the phase is calculated from the moment of the greatest deviation to the right.

The amplitudes of the x- and y-vibrations being respectively $R_0 h \sqrt{\zeta}$ and $\frac{1}{2} R_0 h \sqrt{1-\zeta}$, the vertices of the rectangles, in which the osculating curves are described lie on the circumference of an ellipse with its great axis along the x-axis and having a length of $2\ R_0 h$, and its small axis along the y-axis and having a length of $R_0 h$.

Now ζ changes its value between ζ_1 and ζ_2, so the rectangles in which the osculating curves are described also lie between two extremes.

Moreover as according to (12) to each value of ζ a value of $\cos \varphi$ belongs all osculating curves may now be constructed.

It follows from (13) that for the extreme values of ζ we find $\sin \varphi = 0$; so in the extreme rectangles parabolae are described.

The distance from OX of the node of an arbitrary osculating curve is $-\frac{R_0 h K}{2\zeta}$, from which it is evident that the nodes and also the vertices of the parabolae lie all on the same side of O lying below O for positive values of K (see fig. 2).

Envelope of the osculating curves.

§ 8. If we perform the elimination of t and φ from:

$$x = R_0 h \sqrt{\zeta}\, cos\, n_1 t, \quad y = \frac{1}{2} R_0 h \sqrt{1-\zeta}\, cos\,(2n_1 t - \varphi) \text{ and}$$

$$\zeta \sqrt{1-\zeta}\, cos\, \varphi = K,$$

we find for the equation of the osculating curves with ζ as parameter:

$$\zeta^2 (X^2 + Y^2) + \zeta (KY - X^2 - X^4) + \left(\frac{1}{4} K^2 - 2 K X^2 Y + X^4\right) = 0;$$

where for the sake of a simplified notation is put:

$$X \text{ for } \frac{x}{R_0 h}, \qquad Y \text{ for } \frac{y}{R_0 h}.$$

Thus the envelope has as equation

$$4 (X^2 + Y^2)\left(\frac{1}{4} K^2 - 2 K X^2 Y + X^4\right) - (K Y - X^2 - X^4)^2 = 0.$$

After reduction and division by X^2 (the Y-axis is the locus of the nodes) it may be written:

$$(K - 4 Y^3 - 3 X^2 Y + Y)^2 = (X^2 + 4 Y^2 - 1)^3 (X^2 + Y^2),$$

or if we solve K:

$$K = -(Y \pm \sqrt{X^2 + Y^2}) + (Y \pm \sqrt{X^2 + Y^2})^3.$$

Putting

$$Y \pm \sqrt{X^2 + Y^2} = \frac{K}{U},$$

it passes into

$$K = -\frac{K}{U} + \frac{K^3}{U^3},$$

$$U^2 (1 - U) - K^2 = 0.$$

Now this cubic equation has the same coefficients as (14), so it also has the same roots. So the envelope is degenerated into the 3 parabolae having as equations:

$$U = \zeta_1 \quad , \quad U = \zeta_2 \quad , \ldots \quad U = -\lambda;$$

which after reduction and reintroduction of x and y take the form of:

$$2 \frac{y}{R_0 h} - \frac{\zeta_1}{K} \cdot \frac{x^2}{R_0^2 h^2} + \frac{K}{\zeta_1} = 0 \qquad \zeta_1 \textit{ parabola},$$

$$2 \frac{y}{R_0 h} - \frac{\zeta_2}{K} \cdot \frac{x^2}{R_0^2 h^2} + \frac{K}{\zeta_2} = 0 \qquad \zeta_2 \textit{ parabola},$$

$$2 \frac{y}{R_0 h} + \frac{\lambda}{K} \cdot \frac{x^2}{R_0^2 h^2} - \frac{K}{\lambda} = 0 \qquad \lambda \textit{ parabola}.$$

The parabolae are confocal and have O as focus. When K is positive the ζ_1 and the ζ_2 parabolae have their openings turned upwards,

the λ parabola has its opening turned downwards (this case is represented in fig. 2, where besides some osculating curves the enveloping parabolae are also given).

Special cases.

§ 9. At the close of § 5 we saw that two special cases may occur, viz. when $K=0$ and when $K=\pm\frac{2}{9}\sqrt{3}$.

A. $K=0$. We deduce from the relation

$$\zeta\sqrt{1-\zeta}\cos\varphi=K$$

three possibilities:

1. $\zeta=0$. The movement remains confined to the YZ-plane.

2. $\zeta=1$. The movement remains confined to the XZ-plane. This form of motion however proves to be impossible when $x\neq 0$ and $y=0$ is substituted in (5).

3. $\cos\varphi=0$, therefore $\varphi=\frac{\pi}{2}$ or $\varphi=\frac{3\pi}{2}$ invariably. The osculating curves have their nodes at O. The form of movement approaches asymptotically to a motion in the YZ-plane. What becomes of the enveloping parabolae has been represented in fig. 3, in which some osculating curves have been drawn too.

B. $K=\pm\frac{2}{9}\sqrt{3}$. Then $\zeta_1=\zeta_2=\frac{2}{3}$, $\lambda=\frac{1}{3}$. Now $\cos\varphi=\pm 1$ invariably, thus $\varphi=0$ or $\varphi=\pi$. The same parabola is continuously described, in which also the ζ_1 and ζ_2 parabolae have coincided. (Fig. 4). When K undergoes a slight change, ζ_1 and ζ_2 fall close together. So this form of movement is stable.

$$S=3\ ,\ \frac{\varrho}{n_1}\ \textit{is of order}\ \frac{h}{R_1}.$$

§ 10. The expansions in series written down by Prof. KORTEWEG lose for $S=3$ their convergency as soon as $\frac{h}{R_1}$ passes into order $\frac{\varrho}{n_1}$ (page 18 of his paper) or i. o. w. as soon as $\frac{\varrho}{n_1}$ sinks into order $\frac{h}{R_1}$.

We shall now discuss this case.

We again take as first approximation:

$$x=\frac{\sqrt{\alpha_1}}{n_1}\cos(n_1 t+2n_1\beta_1),$$

$$y=\frac{\sqrt{\alpha_2}}{2n_1}\cos(2n_1 t+4n_1\beta_2);$$

and we must investigate what form the function R now assumes.

As we have supposed that

$$2n_1 = n_2 + \varrho\,,$$

the terms of the order h in the equations of motion would become

$$\ddot{x} + n_1^{\,2}\, x$$

and

$$\ddot{y} + (2n_1 - \varrho)^2\, y.$$

Because $\frac{\varrho}{n_1}$ is of order $\frac{h}{R_1}$ and we take no terms of higher order than h^2 in the equations, we may write for the latter:

$$\ddot{y} + 4n_1^{\,2}\, y - 4n_1\, \varrho y.$$

If we thus take the above expression for x and y as first approximation, then we must admit in the function R besides the term $-d_2\, x^2 y$ also a term $2n_1\, \varrho y^2$.

In the expression

$$-d_2\, x^2 y + 2n_1\, \varrho y^2$$

we substitute the above expressions for x and y and omit the terms containing t explicitly. In this way we arrive at:-

$$R = -\frac{d_2}{N^3}\, a_1 \sqrt{a_2}\, \cos\varphi + \frac{\varrho}{2N}\, a_2\,;$$

where again N is put for $n_2 + \varrho = 2n_1$.

The equations which serve to determine the α's and β's become:

$$\frac{d\alpha_1}{dt} = 2N\, m_1\, a_1\, a_2^{\frac{1}{2}} \sin\varphi\,,$$

$$\frac{d\alpha_2}{dt} = -2N\, m_1\, a_1\, a_2^{\frac{1}{2}} \sin\varphi\,,$$

$$\frac{d\beta_1}{dt} = m_1\, a_2^{\frac{1}{2}} \cos\varphi\,,$$

$$\frac{d\beta_2}{dt} = -\varrho' h + \tfrac{1}{2}\, m_1\, a_1\, a_2^{-\frac{1}{2}} \cos\varphi\,;$$

where

$$m_1 = \frac{d_2}{N^3} \quad , \quad \varrho' = \frac{\varrho}{2Nh}.$$

We again see that

$$\frac{d\alpha_1}{dt} + \frac{d\alpha_2}{dt} = 0,$$

so

$$a_1 + a_2 = constant;$$

for which reason we put:

$$\alpha_1 = \frac{1}{4} R_0^2 N^2 h^2 \zeta \quad , \quad \alpha_2 = \frac{1}{4} R_0^2 N^2 h^2 (1 - \zeta).$$

Further we have according to § 4 as an integral of the system:

$$-\frac{d_2}{N^3} \alpha_1 \sqrt{\alpha_2} \cos \varphi + \frac{\varrho}{2N} \alpha_2 = \text{constant}.$$

Introducing ζ, it becomes

$$\zeta \sqrt{1-\zeta} \cos \varphi - \varrho'' (1-\zeta) = K;$$

where K is a constant and

$$\varrho'' = \frac{\varrho N}{d_2 R_0 h}.$$

In the same way as this was done for the case $\varrho = 0$ we may write down the differential relation between ζ and t and find x and y in the way indicated there as functions of the time; they get quite the same form as for $\varrho = 0$ [1]).

In general ζ keeps changing periodically between two limits ζ_1 and ζ_2; ζ_1 and ζ_2 being the positive roots of

$$\zeta^2 (1-\zeta) - \{K + \varrho'' (1-\zeta)\}^2 = 0.$$

Yet there is a considerable difference between the cases $\varrho = 0$ and ϱ of order h.

§ 11. We notice this difference most distinctly when we represent the relation established between ζ and φ in polar coordinates.

If we put

$$\varrho''' = -\varrho'',$$

then we find:

$$\cos \varphi = \frac{K - \varrho''' + \varrho''' \zeta}{\zeta \sqrt{1-\zeta}}.$$

We take φ as polar angle, $\sqrt{1-\zeta}$ as radius vector and we investigate the site and shape of the curves for positive values of ϱ''' and for all possible values of K.

For $K = \varrho'''$ there is degeneration into the circle $\zeta = 0$ and a straight line normal to the origin of the angles at a distance ϱ''' from pole O_1. We have two cases now: $\varrho''' < 1$ and $\varrho''' > 1$.

$\varrho''' < 1$. Let us now investigate the shape of the curves for different values of K. For $K > \varrho'''$ they lie to the left of the straight line just mentioned, for increasing value of K they contract more and more until for the maximal value of K, belonging to a certain value of

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[1]) Vide Chapter V of my dissertation.

ϱ''' we get an isolated point. If $0 < K < \varrho'''$ the curves surround point O_1; if $K = 0$ we have a curve through O_1, for $K < 0$ they lie to the left of O_1; for the minimal value of K we again get an isolated point (fig. 5).

For increasing values of ϱ''' the straight line separating the domains $K > \varrho'''$ and $K < \varrho'''$ moves to the right. The domain $K > \varrho'''$ becomes smaller and vanishes for $\varrho''' = 1$. For $\varrho''' \geq 1$ we therefore have curves surrounding O_1 and curves to the left of O_1 only. When ϱ''' increases still more the remaining isolated point approaches to O_1 and the curves farther from O_1 approach to circles.

For $\varrho = 0$ we had (with the exception of the special case $K = 0$) only curves to the right of O_1, and curves to the left of O_1. For ϱ of order h we have moreover curves around O_1, which are even more frequent for great values of $\frac{\varrho}{h}$:

The curves around O_1 point to a form of motion, where φ takes all values, the nodes of the osculating curves lie then above as well as below the point O of fig. 2; the osculating parabolae have their openings turned to opposite sides.

That for increasing values of ϱ''' the curves in general begin to resemble circles more and more, indicates that ζ is about constant; it changes between narrow limits.

This also appears in this way. From (16) we deduce:

$$K - \varrho'''(1 - \zeta_1) = \pm \zeta_1 \sqrt{1 - \zeta_1},$$

$$K - \varrho'''(1 - \zeta_2) = \pm \zeta_2 \sqrt{1 - \zeta_2}.$$

By subtraction we find:

$$\zeta_2 - \zeta_1 = \frac{\pm \zeta_2 \sqrt{1 - \zeta_2} \mp \zeta_1 \sqrt{1 - \zeta_1}}{\varrho'''}.$$

For greater values of ϱ''' we find $\zeta_2 - \zeta_1$ becoming very small. In this way we approach the general case where there is no question about relation.

§ 12. How the transition to this general case takes place is also clearly evident from the limitation of the domain of motion, which limitation we find by determining the envelope of the osculating curves. In the same way as this was done for the case $\varrho = 0$, we find that the envelope degenerates into three parabolae, of which the equations are:

$$2\frac{y}{R_0 h}+\frac{K-\varrho'''}{\zeta_1}+\varrho'''=\frac{\zeta_1}{K-\varrho'''}\cdot\frac{x^2}{R_0^2h^2} \qquad \zeta_1 \textit{ parabola},$$

$$2\frac{y}{R_0 h}+\frac{K-\varrho'''}{\zeta_2}+\varrho'''=\frac{\zeta_2}{K-\varrho'''}\cdot\frac{x^2}{R_0^2h^2} \qquad \zeta_2 \textit{ parabola},$$

$$2\frac{y}{R_0 h}-\frac{K-\varrho'''}{\lambda}+\varrho'''=-\frac{\lambda}{K-\varrho'''}\cdot\frac{x^2}{R_0^2h^2} \qquad \lambda \textit{ parabola}$$

The points of intersection of the λ parabola with the ζ_1 and ζ_2 parabolae lie again on the ellipse having R_0h and $2R_0h$ as axes. The parabolae are confocal; the focus lies on the y-axis at the height of $-\frac{1}{2}R_0h\,.\,\varrho'''$. In fig. 6^a, 6^b, 6^c we find those parabolae (and also the osculating parabolae) corresponding to the cases $\varrho'''<1$ and $K \gtreqless \varrho'''$.

In fig. 7 we see how the limitation approaches more and more to a rectangle for increasing ϱ'''.

The ζ_1 and ζ_2 parabolae coincide for maximal and minimal K.

Arbitrary mechanism with 2 degrees of freedom for which $S=3$.

§ 13. Let q_1 and q_2 be the principal coordinates of the mechanism; they remain during the movement of order h and are zero in the position of equilibrium.

The kinetic energy T and the potential energy U may be written:

$$T=\frac{1}{2}\dot{q}_1^{\,2}+\frac{1}{2}\dot{q}_2^{\,2}+T_3\,; \qquad U=\frac{1}{2}(n_1^{\,2}q_1^{\,2}+n_2^{\,2}q_2^{\,2})+U_3,$$

where T_3 and U_3 are expressions in whose terms h appears at least to the 3rd degree.

Let us write down the terms of order h^2 in T_3:

$$T_2=\frac{1}{2}(aq_1\dot{q}_1^{\,2}+bq_2\dot{q}_1^{\,2}+2\,cq_1\dot{q}_1\dot{q}_2+2\,dq_2\dot{q}_1\dot{q}_2+eq_1\dot{q}_2^{\,2}+fq_2\dot{q}_2^{\,2})+\cdots$$

As far as and inclusive of the terms of order h^2 the equations of LAGRANGE now become:

$$\left.\begin{aligned}\ddot{q}_1+n_1^{\,2}q_1=&-\frac{1}{2}a\dot{q}_1^{\,2}-aq_1\ddot{q}_1-bq_2\ddot{q}_1-b\dot{q}_1\dot{q}_2-cq_1\ddot{q}_2-dq_2\ddot{q}_2+\\&+\left(\frac{1}{2}e-d\right)\dot{q}_2^{\,2}-\frac{\partial U_2}{\partial q_1},\\ \ddot{q}_2+n_2^{\,2}q_2=&\left(\frac{1}{2}b-c\right)\dot{q}_1^{\,2}-cq_1\ddot{q}_1-dq_2\ddot{q}_1-e\dot{q}_1\dot{q}_2-eq_1\ddot{q}_2-fq_2\ddot{q}_2+\\&-\frac{1}{2}f\dot{q}_2^{\,2}-\frac{\partial U_2}{\partial q_2}.\end{aligned}\right\}$$

In case the relation $n_2 = 2n_1$ is strictly satisfied or nearly so, the disturbing terms are:

in the first equation those with $q_2\ddot{q}_1$, $\dot{q}_1\dot{q}_2$, $q_1\ddot{q}_2$, q_1q_2,

„ „ second „ „ „ „ $\dot{q}_1^2$, $q_1\ddot{q}_1$, q_1^2.

If at first approximation we try to satisfy the equations by:

$$q_1 = Ah\cos(n_1t + \lambda) \quad , \quad q_2 = Bh\cos(n_2t + \mu)$$

where A, B, λ and μ are functions of t, however in such a manner that $\dot{A}$, $\dot{B}$, $\dot{\lambda}$, $\dot{\mu}$ are of order h or smaller, we may replace in the second member of the equations:

$\dot{q}^2_1$ by $n_1^2(A^2h^2 - q^2_1)$, $\dot{q}_2^2$ by $n_2^2(B^2h^2 - q_2^2)$,

$\ddot{q}_1$ by $-n_1^2q_1$, $\ddot{q}_2$ by $-n_2^2q_2$.

If we take this into account for the disturbing terms and if we omit the non-disturbing terms, the equations become:

$$\left.\begin{aligned} \ddot{q}_1 + n_1^2q_1 &= (bn_1^2 + cn_2^2 + 2p)\,q_1q_2 - b\dot{q}_1\dot{q}_2 \\ \ddot{q}_2 + n_2^2q_2 &= \left(2cn_1^2 - \frac{1}{2}bn_1^2 + p\right)q_1^2. \end{aligned}\right\}$$

The terms $2pq_1q_2$ in the first equation and pq^2_1 in the second originate from a term $-pq^2_1q_2$, appearing in U_3.

To get rid of the term with $\dot{q}_1\dot{q}_2$ we use the new variable q' so that:

$$q'_1 = q_1 + \frac{1}{2}bq_1\,q_2.$$

Then:

$$\ddot{q}'_1 = \ddot{q}_1 + \frac{1}{2}b\ddot{q}_1\,q_2 + \frac{1}{2}bq_1\,\ddot{q}_2 + b\dot{q}_1\,\dot{q}_2 =$$

$$= \ddot{q}_1 + b\dot{q}_1\,\dot{q}_2 - \frac{1}{2}b(n_1^2 + n_2^2)\,q_1\,q_2.$$

Therefore:

$$\ddot{q}_1 + b\dot{q}_1\,\dot{q}_2 = \ddot{q}'_1 + \frac{1}{2}b(n_1^2 + n_2^2)\,q_1\,q_2.$$

The equations now pass into:

$$\left.\begin{aligned} \ddot{q}'_1 + n_1^2\,q'_1 &= \left(bn_1^2 + cn_2^2 - \frac{1}{2}bn_2^2 + 2p\right)q'_1\,q_2, \\ \ddot{q}_2 + n_2^2\,q_2 &= \left(2cn_1^2 - \frac{1}{2}bn_1^2 + p\right)q'^2_1. \end{aligned}\right\}$$

For we may replace in the second members q_1 by q_1', as their difference is of order h^2.

H. J. E. BETH. "The oscillations about a position of equilibrium where a simple linear relation exists between the frequencies of the principal vibrations." (Second part).

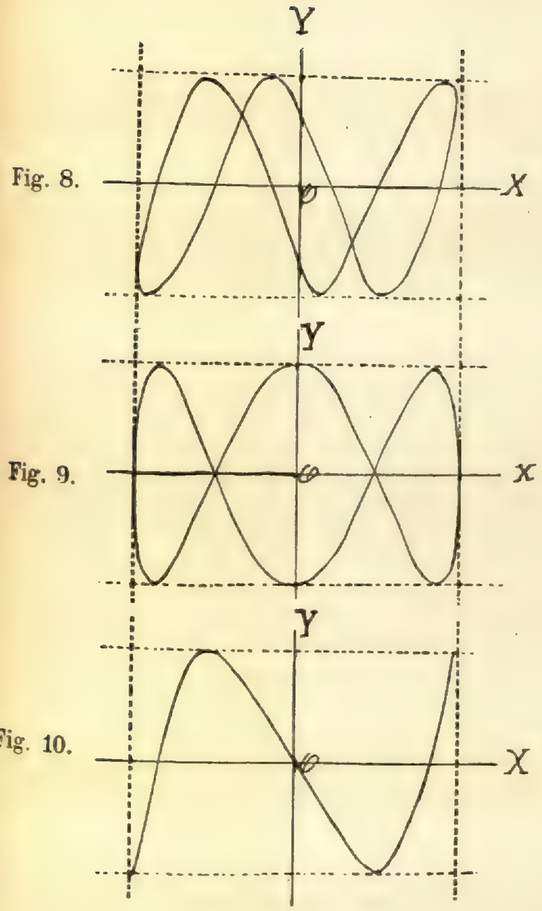

Fig. 8.

Fig. 9.

Fig. 10.

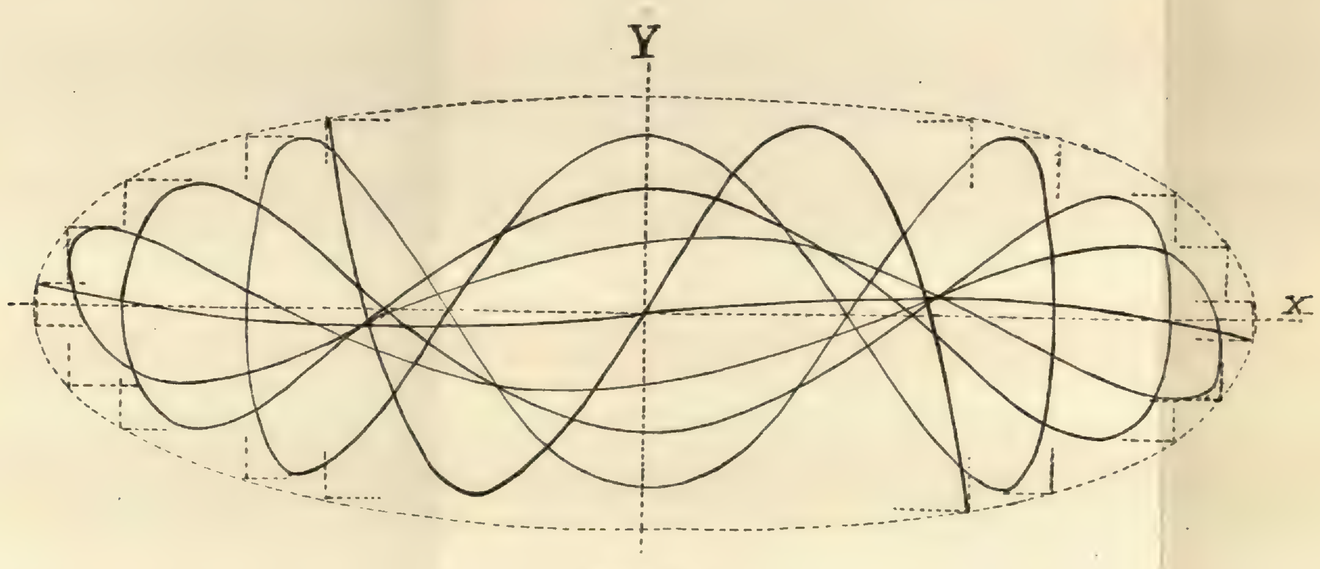

Fig. 11.

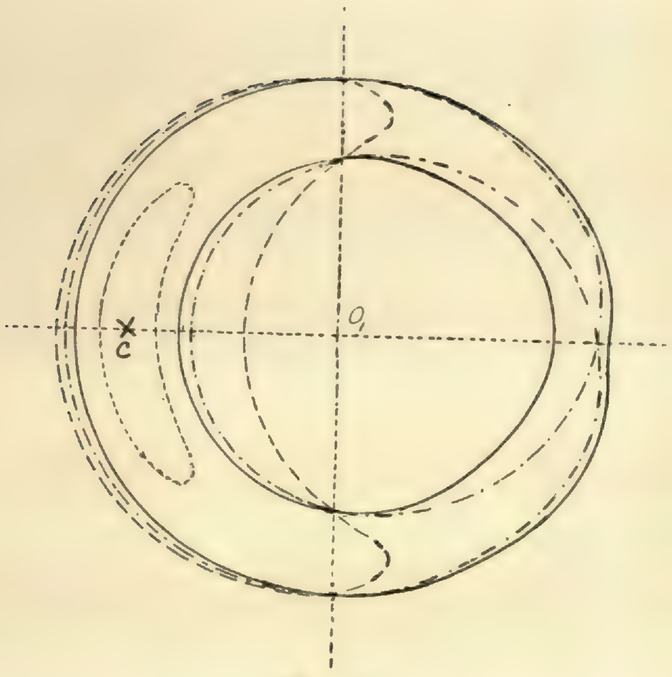

Fig. 12.

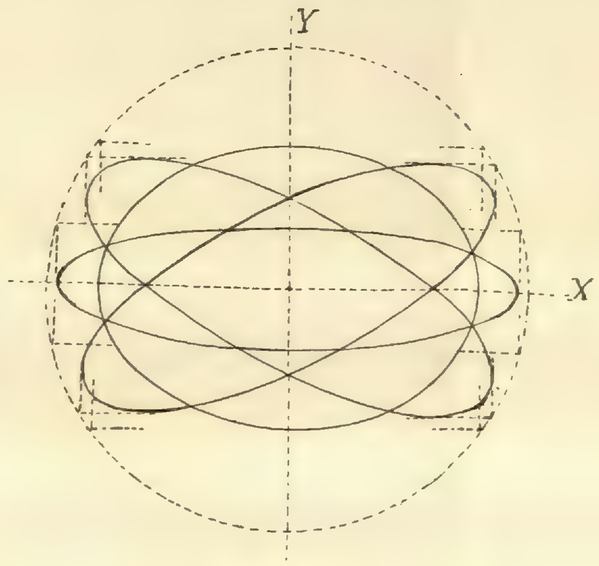

Fig. 13.

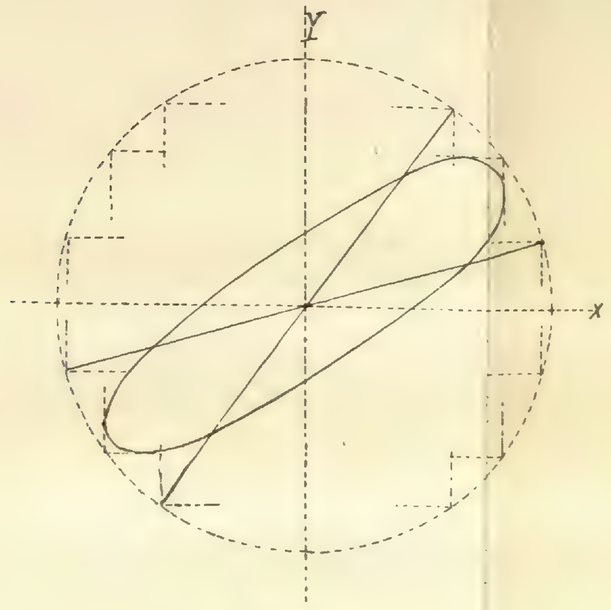

Fig. 14.

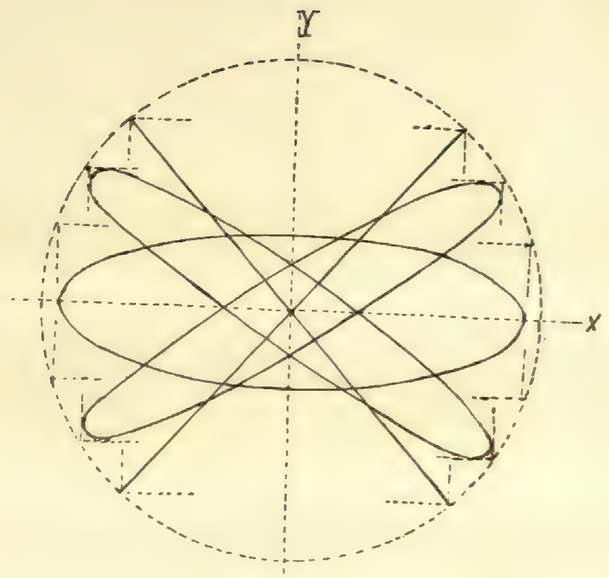

Fig. 15.

Let us now suppose n_2 to be $= 2n_1$; then we get:

$$\ddot{q}'_1 + n_1^2 q'_1 = (4\,c\,n_1^2 - b\,n_1^2 + 2\,p)\,q'_1\,q_2,$$

$$\ddot{q}_2 + n_2^2 q_2 = (2\,c\,n_1^2 - \frac{1}{2}\,b\,n_1^2 + p)\,q'^2_1.$$

So we find:

$$\begin{cases} \ddot{q}_1' + n_1^2 q'_1 + 2\,d_2\,q'_1\,q_2 = 0, \\ \ddot{q}_2 + n_2^2 q_2 + d_2\,q'^2_1 = 0; \end{cases}$$

where

$$d_2 = -\,2\,c\,n_1^2 + \frac{1}{2}\,b\,n_1^2 - p$$

The equations determining the first approximation have exactly the same form as those found in § 4. What was formerly deduced for the simple mechanism holds consequently, if $n_2 = 2n_1$, for an arbitrary mechanism with two degrees of freedom in such a sense that the horizontal projection of the point moving over the surface may be regarded as the representative point for the arbitrary mechanism.

We finally observe that any mechanism for which

$$-\,2cn_1^2 + \tfrac{1}{2}bn_1^2 - p = 0$$

is not sensitive for the relation $n_2 = 2n_1$. So this is the condition requisite to make the mechanism for $n_2 = 2n_1$ a mechanism of exception in the sense indicated by Prof. KORTEWEG (§ 26 of his paper).

Mechanisms of exception therefore are among others the symmetrical mechanisms (§ 31 of that paper); for here c, b, and p are all equal to zero.

Microbiology. — *"Viscosaccharase, an enzyme which produces slime from cane-sugar"*. By Prof. Dr. M. W. BEIJERINCK.

The emulsion reaction.

Many spore-producing and a few non spore-producing bacilli, cause, when growing in presence of cane-sugar or raffinose on neutral or feebly alkaline agarplates, a very peculiar "colloidreaction", which is also valuable for the diagnosis of these bacteria. This reaction consists in the formation, in and also on the surface of the agar around the colonies or streaks, of a liquid "precipitate", i. e. an emulsion, which can best be recognised in transmitted light, and at the same time in a swelling of the agar caused by the increase of volume produced by the emulsion.

The emulsion consists of drops (see plate) of different size, mostly very small, but sometimes growing to 0,2 mm. so that they may

be distinguished with a magnifying glass. At feeble magnification they might be taken for droplets of oil suspended in the agar, but as strong sulphuric acid dissolves these drops immediately, and a feebler acid more slowly, there can be no question of oil or fat.

Characteristic for the reaction is that it can only be distinctly observed in agar but imperfectly in gelatin. In the agar the process is impeded when acid is produced by the microbes. Thus bouillon-agar, yeastwater-agar, and wort-agar with cane-sugar can well be used, but the emulsion is more distinctly formed in agar with mixtures of substances that prevent the acidification, to which cane-sugar is so very apt. For that reason nitrates as nitrogen-food are especially favourable, as the withdrawing of nitrogen then necessarily must produce an alkali, while for example ammonium-salts, used as source of nitrogen, must promote the acid reaction.

A good experiment to produce the emulsion is the following: A plate is prepared of the composition: tapwater, 2 °/₀ of agar, 2 °/₀ of cane-sugar, 0,02 °/₀ KNO_3 and 0,02 °/₀ K_2HPO_4. Nitrogen food may also be quite left out, so agar-plates with 10 °/₀ of cane-sugar and bikalium-phosphate only, are very well fit to demonstrate the emulsion with *Azotobacter* and the hereafter mentioned *Bacillus emulsionis*. The quantity of cane-sugar can vary between 0.1 °/₀ and 50 °/₀ without much difference in the result.

After the solidifying of the agar-plate and the removal of the adhering water, soil-bacilli are dispersed, obtained by shaking some garden-soil with water, and heating it a few minutes at 70° to 80° C. in order to kill the not sporulating microbes, Then the water is poured over the plate and allowed to flow off. The adhering germs, for so far they live, are nothing but spores of bacilli, which can germinate at 30° C.

After one or two days the colonies become visible and simultaneously the emulsion around some of them; the majority does not produce the emulsion.

Cane-sugar may be replaced by raffinose, which acts in the same way; but glucose, levulose, mannose, galactose, lactose, maltose, trehalose, melibiose, mannite, inulin, dextrin and xylose, do not give the emulsion.

The emulsion is distinct round the colonies of *Bacillus mesentericus vulgatus* (see plate Fig. 1), *B. megatherium* and a not yet described soil-bacillus, commonly also found in cane-sugar itself, recognisable by its small terminal spores, which may be called *Bacillus emulsionis* and whose transparent colony is likewise given on the plate (Fig. 2). The emulsion is wanting in *B. subtilis*, *B. mycoides*, *B.*

polymyxa, *B. nitroxus*, *B. sphaerosporus*, *B. luteus*, besides in the anaërobes *Granulobacter butylicum*, *Gr. saccharobutyricum* and *Gr. pectinovorum*.

The moulds, the various yeast species, even those which invert cane-sugar, besides all species of *Streptothrix*, and most of the non-spore producing bacteria, do not produce the emulsion either.

An exception to the last rule makes the non-spore producing *Azotobacter chroococcum*, which on plates of 2 % of agar, 2 to 10 % cane-sugar, and 0,02 % K_2HPO_4, in water, gives a strong emulsion, which extends to a large distance round the colonies; later, in their vicinity, perhaps by the influence of a specific enzyme or an acid it vanishes, while near the colonies of the soil-bacilli the emulsion is permanent. With the exception of *B. chroococcum* the other forms of *Azotobacter* do not produce the emulsion. From cultures of *Azotobacter*, prepared with garden soil and destined for the absorption of free nitrogen, a species related to *B. radiobacter* can be obtained, which produces no spores, but does also give a strong emulsion.

Ane mulsion, from a physical view analogous but quite different by the manner in which it takes rise, was described by me on another occasion[1]). It appears when a 10% solution of gelatin in water is boiled with a 10% solution of soluble starch, or with a 2% agar-solution. Even by boiling the two watery solutions do not mix, which, of course, is also the case after solidifying. This reposes evidently on the fact that here two colloidal solutions are brought together, which cannot diffuse and whose emulsionated droplets constantly have a positive surface-tension with regard to each other. The same explanation must hold good for the emulsion formed by the viscosaccharase with regard to the agar, and as I may add, to culture-liquids wherein *Bacillus emulsionis* produces the emulsion also.

The emulsion is produced by an enzyme.

If from the emulsion field round a colony a small piece of agar is cut out, without touching the colony, and placed on an other cane-sugar-agar-plate, the emulsion itself does not diffuse out of it, but into the plate, a substance goes over, which produces the emulsion again and with regard to the quantities used rather strongly. This points with certainty to the presence of an enzyme as the cause of the emulsion, an enzyme which must have the property of moving through the agar by diffusion. This agrees perfectly well with the

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[1]) Centralbl. f. Bacteriologie 2te Abt. Bd. 2, p. 627, 1896.

origin of the emulsion round the colonies, for a substance which is evidently insoluble in the agarplate, can only be found at the place where it is produced. This substance having in our case the nature of a plant slime, the enzyme may be called *viscosaccharase*.

The enzyme is prepared by filtering a culture of *B. mesentericus vulgatus* and precipitating the filtrate with alcohol, whereby, of course, other enzymes formed by this bacterium such as diastase, and also the slime substance itself, are precipitated. Whether to the enzymes, present in this mixture invertase must be reckoned, which is usually considered as a secretion-product of *B. mesentericus*, has become doubtful by the discovery of the viscosaccharase, at whose action, as will be seen below, together with the slime, the production of a reducing sugar is stated.

Even in presence of chloroform the emulsion reaction takes rise on cane-sugar agar-plates through the enzyme produced from the *mesentericus* cultures, without anything being perceived of the development of the germs of *B. mesentericus* itself, which may be still present after filtering and precipitating.

It is not difficult to prepare plates of any size containing the emulsion everywhere, and fit for experiments to demonstrate by what influences it may disappear.

To this end the required culture-agar is mixed before solidifying with a not too large number of germs, for example of *B. emulsionis*, and then placed one or two days in the thermostat ; when the plate becomes quite turbid by the emulsion, the sugar is washed out and it is ready for the experiment. A drop of dilute acid thereon rapidly produces a clear space.

At the action of viscosaccharase, besides the slime a reducing sugar is found.

When small pieces of agar containing the emulsion are introduced into an experiment-tube and cautiously warmed with a little FEHLING's copper solution, a strong reduction is seen, which does not take rise with the same sugar-agar if the emulsion is wanting.

The question arose whether this reaction should be ascribed to the slime itself, or if at the same time, through the viscosaccharase, or in another way, some other reducing substance is formed. Therefore small pieces of the agar containing the emulsion were washed out with water, whereby the slime, which cannot diffuse from the agar into the water, remains behind, but the reducing power of the agar is lost, whilst the water used for the washing becomes itself strongly

Fig. 1. Bacillus mesentericus

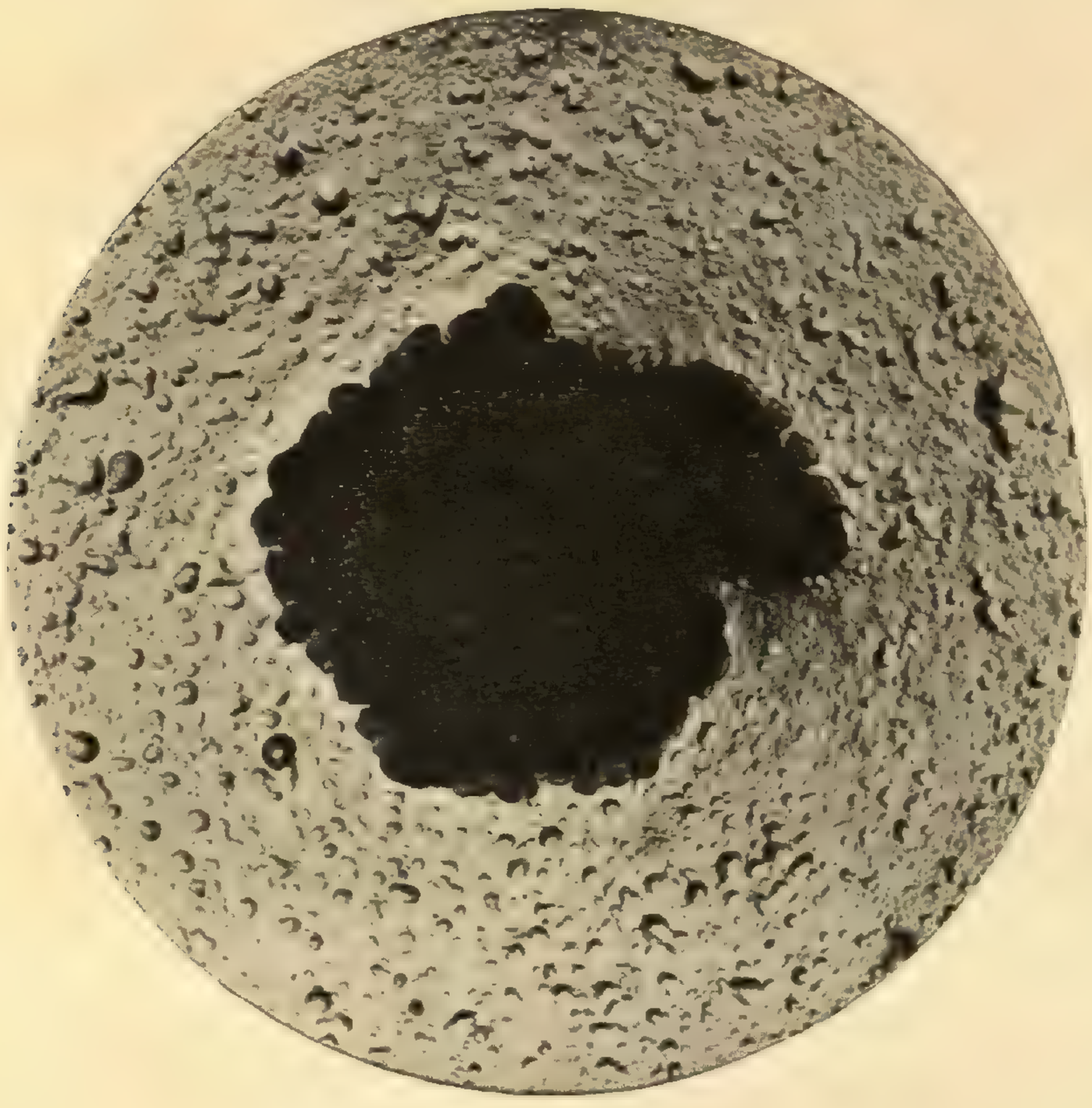
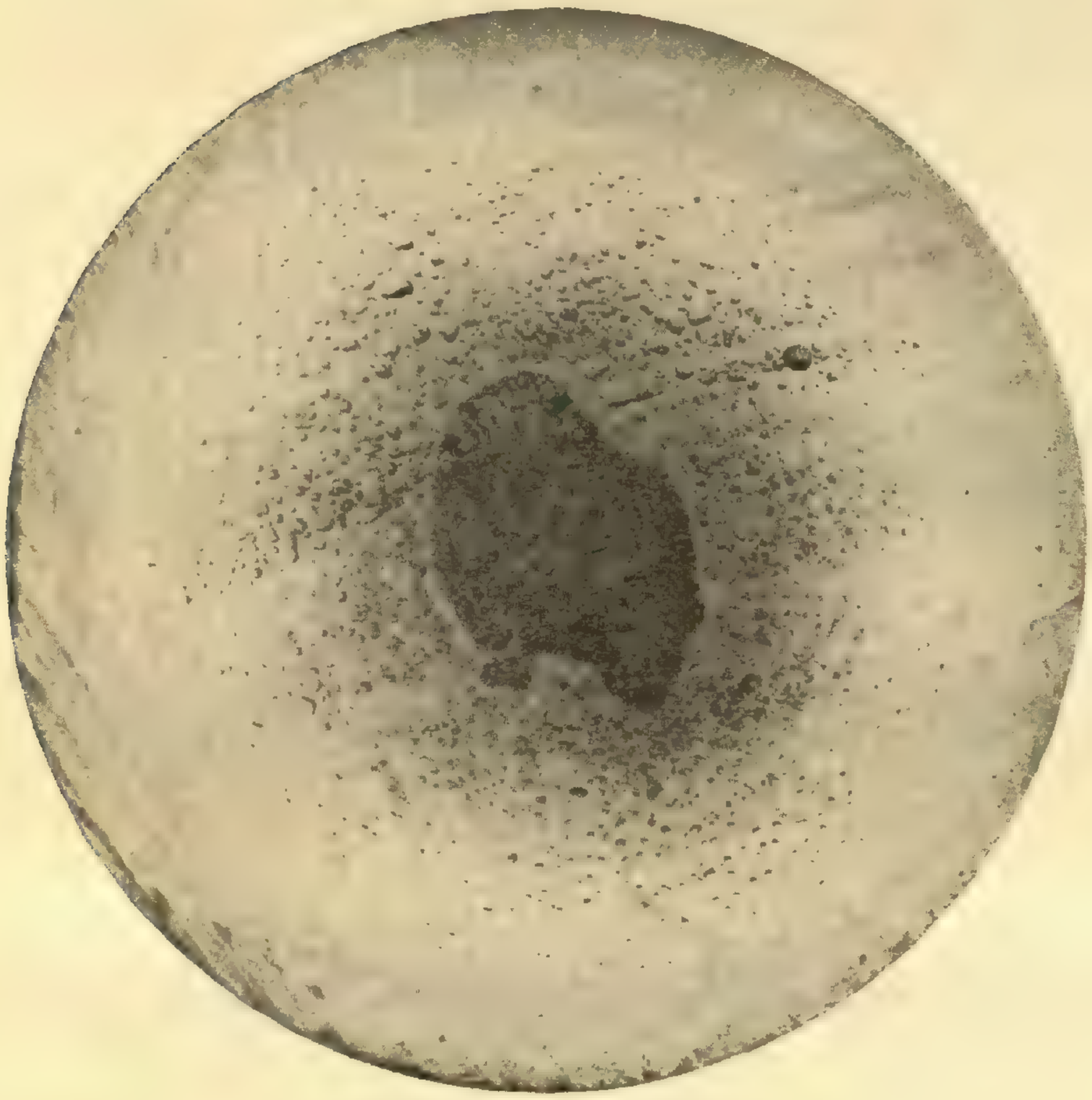


Fig. 2. Bacillus emulsionis.



The emulsion-reaction.

reducing. Hence it is sure that at the "emulsion reaction", together with the non-reducing slime, an easily diffusing and reducing substance (probably a sugar) is formed. The chemical composition of this substance is still unknown, just like that of the slime itself.

The possibility exists that the reducing substance is invert-sugar produced by invertase, which latter enzyme then should always accompany the viscosaccharase. Decisive experiments on this subject in progress.

Viscosaccharase is a synthetically acting enzyme.

As to the nature of the slime it must be accepted that its molecules are much larger than those of cane-sugar, else it would not be clear why the slime cannot diffuse through the agar, which cane-sugar does very easily. Viscosaccharase must therefore be a synthetically acting enzyme. This circumstance suggests a relation between the slime and "dextran" [1]). This is, however, a substance forming the cell-wall of the concerned microbes, which substance may spread in water, and even to some extent diffuse into agarplates, but is not the product of an exo-enzyme, i.e. of an enzyme able to leave the bacterial body and act outside of it like the viscosaccharase. In relation to this it is not astonishing that "dextran" can very well originate from glucose and some other sugars, which do not produce the emulsion.

Very remarkable is the fact that all the hitherto examined bacteria which show the emulsion-phenomenon, are able, at definite culture-conditions, for example on cane-sugar gelatin, when no emulsion is produced, to form non-diffusing "dextran", by which their colonies then become visible on the plates as large transparent drops. This also points to a narrow relation between the two phenomena and leads to the conclusion that the drops of the emulsion must be identic with, or related to dextran.

Perhaps by further research modifications of viscosaccharase will prove to exist, which also act on glucose and other sugars and from these may form "dextran", but which cannot leave the body, or rather the cell-wall of the microbes, and must be considered as endo-enzymes whose product, which itself does not diffuse, cannot be found beyond the limits of the colony.

If in accordance with my expectation, the emulsion is really brought about by "dextran", then light will be thrown on the formation of the wall-substances of plant cells in general; for there is no doubt

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[1]) C. Scheibler, Zeitschr. d. Vereins für Rübenzuckerindustrie, Bd. 24, p. 309, 1874. L. Maquenne. Les sucres et leurs principaux dérivés. p. 745, 1900.

that "dextran" is a modification of cellulose, and the till now not explained secondary changes, observed in so many cell-walls, may then freely be ascribed to the action of specific enzymes, related to the viscosaccharase.

Why the emulsion is distinctly observed in agar, and less easily in gelatin-plates, must probably be explained by the dimension of the molecules of viscosaccharase, which are small enough to enter without much trouble the relatively wide canals of the agar, but too large to pass through the much narrower ones of the gelatin.

Many of the experiments here related I owe to Mr. D. C. J. MINKMAN, assistant in my Laboratory.

EXPLANATION OF THE PLATE.

Fig. 1. Colony of *Bacillus mesentericus vulgatus* on: canal water, 2% of agar, 1% of cane-sugar, 0.02% KNO_3 and 0.02% K_2HPO_4, with emulsion around colony. Magnified 8 times.

Fig 2. Colony of *Bacillus emulsionis* n. sp., on canal water, 2% of agar, 0.1% of cane-sugar, 0.02% $ClNH_4$, 0.02% K_2HPO_4, with emulsion around colony, Magnified 9 times.

Microbiology. — "*Variability in Bacillus prodigiosus.*" By Prof. M. W. BEIJERINCK.

In a former paper [1]) I showed how easily new constant variants of *Bacillus prodigiosus* and other microbes may be obtained. Here follow some further observations, made with the aid of Mr. H. C. JACOBSEN, assistant in my Laboratory.

The keeping constant of the cultures.

The principle on which the keeping constant of *B. prodigiosus* seems to repose is preventing the cultures from becoming alkaline by their own action. Thus, by re-inoculating in quick succession, for instance every 24 hours, into bouillon or on bouillon-agar at 30° C., each form of *Bacillus prodigiosus*, whether the natural or normal form, or a variant obtained from it, remains unchanged probably for an indefinite time.

For the transplantations only very little material must be used and an abundance of food.

If some lactic acid is added, for instance 0,5 to 1.5 cm^3 normal per 100 cm^3 of bouillon, the culture likewise remains unchanged

[1]) Royal Acad. of Sciences 21 Nov. 1900,

after a prolonged series of transports, if these are always carried out before the acid is neutralised by the alkali produced from the bouillon by the bacteria themselves [1]).

Addition of 1 to 2 pCt. of glucose acts in the same manner as free acid, *B. prodigiosus* therefrom producing acid which may rise, if sufficient glucose is added, to 3 to 4 cm³ normal per 100 cm³ of bouillon. As the titre of alkali, originating in the bouillon alone, can amount to 2.5 cm³ N per 100 cm³ of bouillon, and as from 1 pCt. of glucose there results no more than 1.5 to 2 cm³ N of acid, addition of 1 pCt. of glucose is sufficient to prevent variation, if the re-inoculations take place quickly; but not if effected with long intervals, for in the latter case more alkali may result from the bouillon than acid from the glucose.

If to the bouillon so much ammoniumcarbonate or natriumcarbonate is added that the titre of alkali amounts to about 3 cm³ N per 100 cm³ of the medium, *B. prodigiosus* likewise remains constant after repeated inoculations at 30° C., whilst the control culture, without carbonate but for the rest under the same conditions, strongly varies. The same result may be obtained with magnesiumhydrophosphate ($Mg\,H\,PO_4\,.\,2\,H_2O$) to excess; this, however, quickly precipitates, and in order to be active should be used in a bouillon-agarplate or in a thin layer of liquid. In ordinary bouillon-agarplates 1 pCt. of this salt changes entirely into crystals of ammoniummagnesiumphosphate ($Mg\,NH_4\,PO_4\,.\,6\,H_2O$) the plate becoming quite transparent; a plate with 3 to 4 pCt. on the other hand, remains white and turbid.

Although it may be admitted that by these various means the formation of secretion products by the bacteria is prevented, on whose stimulating action the variability probably reposes, yet it, is not clear how this preventing takes place. Evidently substances should be thought of here which, once produced, cannot or only with difficulty leave the bacterial body.

Of the said means quick transplantation is the simplest for always disposing of constant stocks for the experiments.

The origin of the variants in general.

When cultures, placed under favourable nutritive conditions, but for the rest prepared without special precautions, are growing older between 10° and 30° C., they exhibit a certain variability at which, as formerly described (l. c.), variants are thrown off, while beside

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[1]) At 4 cm³ of acid per 100 cm³ of culture liquid the growth of *B. prodigiosus* is slackened, at 9 cm³ it is quite stopped.

these the original form is found unchanged. As by transplantations in rapid succession (and under constant and favourable conditions) no change occurs during thousands of cell-partitions, this variability cannot repose on some law governed by internal causes only, but a particular agency is wanted, which may have its seat within the cells, but which must yet be enacted on by external circumstances.

Although the variability can reveal itself already in an ordinary same well arranged culture, e.g. in bouillon or in maltwort, allowed to stand for a few weeks, yet this process may considerably be accelerated by repeated transplantations, *not* after a very short time, but with longer intervals, for example two days, with cultures kept at 30° C., a not too small quantity of the material for the inoculation being used, e. g. two loops of the platinum thread. After three or four repetitions, so after about a week, the variation can then be in full course, the first culture, left to itself, not yet showing any perceptible change.

This evidently reposes on the following circumstance. The influence which causes the variability in the culture when it gets older, acts in the chosen conditions already after two days. If now a re-inoculation is performed, the germs affected by that influence can increase as well as those that remained normal, whilst by not re-inoculating, thus in the first culture, the non-affected germs are by far more numerous and remain so as the cell-division slackens after the second day, because of want of food. At inoculation after two days there result at each time new modified germs, and those which are modified already, are enabled to augment without losing their modification.

In this explanation it must further be accepted, that a transplantation after two days gives no cause for atavism; for if this were the case, the reverse ought to take place of what is observed: after a week's growth the first culture should be more varied than that which has repeatedly been transplanted, but this is not so. This shows how carefully the variation experiments must be carried out in order not to become obscure.

Particularly the cultures on solid media must very accurately be observed. If these are allowed to stand for some days or weeks without further precautions, then in many cases, even with magnifying glass or microscope no variation at all can be detected, although it is actually going on, commonly to "rose" or "white".

Colony culture then shows that here and there varied germs or groups of such germs must be present, for from the seemingly homogeneous matter large numbers of white and rose variants are

obtained, which prove as constant as the normal form itself. However unchanged colonies, representing the pure stock and producing a material as fit for further experiments as the original culture, lie among the variants.

Experiences afforded by other bacteria seem to prove that the frequent repetition of the thus possible process of selection, produces a form which varies less than the original material. But it is not here the place to enter upon this important fact.

All colony cultures of *B. prodigiosus* are best made on bouillon-agar-plates, which after solidifying have been cautiously dried on a thermostat at circa 40° C. The water which then condenses on the glass cover can easily be removed; if this is neglected, *B. prodigiosus*, which is strongly motile, spreads over the surface of the agar and the colonies coalesce.

I shall now enter into a short discussion of the most important variants.

The obtained variants.

The variants derived from *B. prodigiosus* may be considered as plus- or gain-variants, minus- or loss-variants, and qualitative variants. This is exposed below in the table of descent, which shows the origin of the obtained forms; the qualitative variants (*auratus* and *hyalinus*) are placed on the same line with the normal form, the plus-variants above it, the minus-variants beneath. Hence, the arrows not only denote the descent but also whether the variability reposes on gain or loss of characters, or if it is qualitative. Dotted arrows indicate that atavism has with certainty been observed. The names indicate the chief qualities characterising the variants.

A survey of the variants without regard to their descent precedes; then follows their pedigree, which does not repose on hypothesis, but simply gives the result of the experiments.

The obtained variants are:

1. *Bacillus prodigiosus.* Normal form, isolated from nature [1]).
2. „ „ *roseus* 1.
3. „ „ „ 2.
4. „ „ *albus.*
5. „ „ „ *hyalinus.*
6. „ „ *viscosus.*
7. „ „ „ *albus.*
8. „ „ *auratus.*

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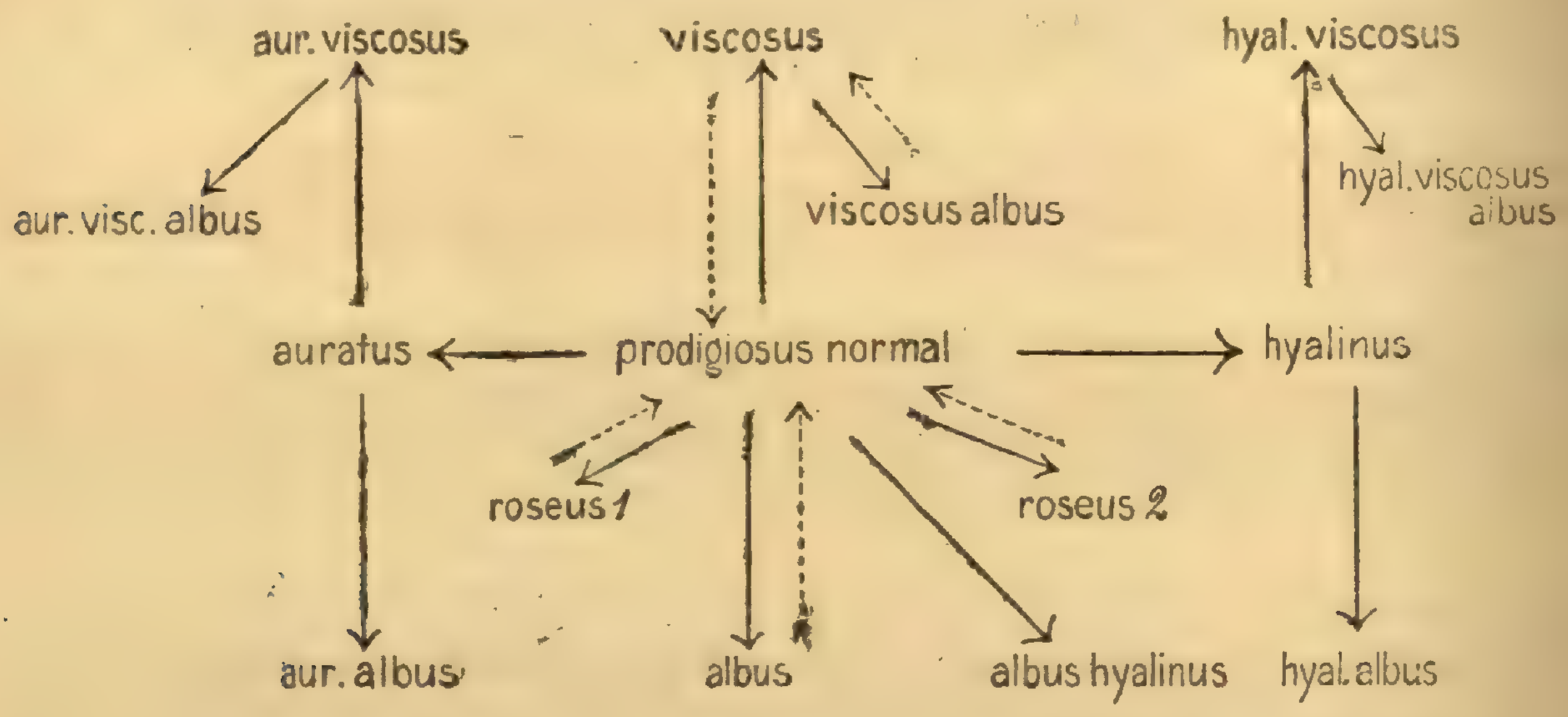
[1]) About 1890 from mouldering bones of a gelatinfactory near Delft.

9. *Bacillus prodigiosus. auratus viscosus.*
10. „ „ „ „ *albus* (= 7 ?)
11. „ „ „ *albus* (= 4?).
12. „ „ *hyalinus.*
13. „ „ „ *viscosus.*
14. „ „ „ „ *albus.*
15. „ „ „ *albus* (= 5?)

The relation and origin of these variants is given in the following table.



The upward arrows denote "gain-variation", the horizontal "qualitative variation", the downward arrows "loss-variation". Dotted arrows signify that atavism has been observed.

The two qualitative colour-variants, *auratus* which is orange-coloured and *hyalinus* of a deep vine-red, vary in a way quite corresponding to the normal form and like this throw off, under the same circumstances, slime-variants and white variants. Besides, the normal form may return by atavism as well from *auratus* and *hyalinus* themselves as from the variants derived from them. In the pedigree table atavism is indicated by dotted arrows for a few of the cases where it has been stated with certainty. But there is no doubt that also the other variants are disposed to atavism.

It should moreover be noted that the *auratus*-variant approaches, at least in colour, the natural variety *Bacillus Kieliensis*, but that the latter possesses a stronger power of fermentation, and produces much gas ($CO_2 + H_2$) from maltwort with dextrose or cane-sugar, the former fermenting only dextrose.

For the rest, *B. Kieliensis* itself, which varies in a way quite analogous to that of the normal form of *prodigiosus* here considered, has not yet been obtained as a variant from the latter,

A new character which may rise in addition to the already existing ones, is the production of a large quantity of slime substance by excessive growth of the cell-wall, which slime may spread through the liquids, and makes the individuals of the colonies on agarplates cohere into one tough mass. From *B. Kieliensis* was even a variant obtained whose colonies appear on the agar plates as a very consistent, almost dry zoogloea, but the analogous variant did not till now arise from the common *prodigiosus*. The *viscosus* (6), derived from the latter, is an ordinary red slime bacterium.

This red-coloured, tough-slimy form, which may be called *B. prodigiosus viscosus*, is no doubt a plus-variant. Its production has been observed under the most different nutritive conditions, between the temperatures 10° (in a cellar) and 30° C., but always and exclusively in liquid media, never on a solid one. The latter circumstance is apparently the reason why the numerous experimenters, who have studied *B. prodigiosus*, have not seen this variant. It is true that SCHEUERLEN [1]) observed that old *prodigiosus*-cultures sometimes turn slimy, but he ascribed it to their becoming alkaline and overlooked that a new constant form was produced.

The only distinct condition which seems different in the liquid cultures compared with the solid, is the access of oxygen. In the depth of the liquid this access must, of course, be very deficient for a long time, or even be entirely lacking, as the upper layers of the culture, which are rich in bacteria, take up all the oxygen. Consequently anaërobiose becomes possible in the depth, which is not the case in cultures lying free on a solid medium, and this partial anaërobiose is apparently the stimulus which induces the formation of the slime variant. That here a rather complex influence and not a direct action must be ascribed to the partial withdrawing of the oxygen, follows from the fact that the culture of *B. prodigiosus* at complete exclusion of air, as in a closed bottle, does not, even with repeated transports, give rise to the slimy variant. At temperatures of about 35° C. this variant is no more formed, although the growth of *prodigiosus* is then still very strong; at 37° the growth slackens or ceases entirely, according to the food.

In the following liquid media the production of the slime variant has with certainty been observed, as well after repeated re-inoculations as after prolonged keeping of one and the same culture at 25° to 30° C.: in broth, in broth with 1 pCt of glucose, in malt-wort, in tap-water with 5 pCt of pure gelatin and 0,02 pCt K_2HPO_4, and in

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[1]) Archiv. für Hygiene. Bd. 26 p. 1.

tap-water with 2 pCt of glucose, 0.5 pCt of asparagine, 0,02 pCt K_2HPO_4, always cultivated at 30° C. and with repeated transports after two days or longer. From this we also recognise that there is no question of a direct influence of the food on the production of the variant.

The *auratus*- and *hyalinus*-variants, also, have only taken rise in liquid cultures, namely in broth and in the glucose-asparagine solution. Moreover, *hyalinus*, which is of a deep vine red, is easily obtained from a solution of pure gelatin in tap-water with 0.02 pCt. K_2HPO_4, after repeated re-inoculations, at 30° C., whereby also *hyalinus viscosus* results.

The colourless or white variants, which only differ from the original form in producing no pigment, should certainly be considered as minus-variants. They are obtained with more ease than the slime variants and, at least as to N° 4, have also been detected by other authors [1]).

Except under the said conditions, apt to keep them constant, all the cultures as well in liquid as on solid media, vary sooner or later towards white. The original form does remain preserved, but a colourless variant is thrown off, which is still more constant than the stock itself.

Not always does one and the same variant result in this case: two uncoloured constant forms, N° 4 and 5 can easily be distinguished if they originate at the same time, and their colonies are on the same agarplate so that they may be compared somewhat magnified. One, *albus hyalinus*, then looks more blueish transparent, the other, *albus*, is more of a cloudy and opake white; under the microscope the former proves to consist of smaller cells than the latter.

The cause of the production of white variants cannot be a more or less abundant access of oxygen, but must probably be sought in a stimulus, exerted by secretion products which remain enclosed in the interior of the cells.

Although the presence of ammoniumcarbonate in the medium (broth-agar), as also cultivation at temperatures higher than 30° C. e.g. at 33° C., prevent pigment production, no hereditary variation at all is caused by these influences. If the thus treated colourless cultures are transported at 20° to 25°, no white variants are obtained from them, but the normal form is found back unchanged, if at least the above mentioned precautions to preserve the constancy of the stock are not neglected.

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[1]) In LEHMANN and NEUMANN's Atlas, 4th Ed. 1907, Table 30, Fig. 3, shows a coloured image of a "pure culture" of *prodigiosus*, consisting of red and white colonies.

When the white variants of the normal form are cultivated at 30° C. in bouillon or in malt-wort, the cultures will, after a few re-inoculations, turn slimy like those of the red normal form itself. Colony culture on bouillonagar proves that white slime variants are thrown off, in the same way as the normal form throws off the red ones. The white slime variants (N°. 7 ? and 14) correspond by the nature of their colonies to the two white forms, *albus* (4) and *albus hyalinus* (5), considered above.

There is still another method to obtain the colourless slime variant from the red one. If this latter is cultivated at 30° in malt-wort or in bouillon, we find after one or two transferrings, each time after two days, and when sown on bouillon-agar, many white slime colonies together with the unchanged red, moreover a considerable number of quite normal, not slimy red colonies, N°. 1, which is to be considered as atavism, but an atavism reposing on the loss of a character. The white slime variant, thus obtained by minus-variation, and found, in the table as N°. 7, seems identic with the one produced by plus-variation from the not slimy white variant, which latter for that reason has not been specially mentioned.

Already in my earlier paper I spoke of rose variants, which so to say, keep the middle between the normal form and the white variant. They may be produced in various ways, for instance, by cultivating the normal form on plates of pure gelatin dissolved in distilled water (H_2O, 10% of gelatin) at room temperature, at which rapid growth and vigorous melting occur. By daily streaking off on a bouillon agarplate the same colony obtained on such pure gelatin, and provided the temperature be kept between 14° and 17° C., we find, on the fifth or sixth day, the first rose variants, either or not with the white, which under these conditions appear later. Two rose variants (table N°. 2 and 3) are easily distinguished, but it is possible that there are many more whose perception is beyond the reach of our observation. In any case, it is a fact that the character: "the faculty of producing pigment", is divisible in many ways. The hereditary constancy of at least one of these rose variants proved not to differ from that of the normal form.

Another method to obtain rose variants is cultivation of the normal form in bouillon, which by evaporation has been reduced to a threefold concentration. After a single transport already, a large number of rose variants (3) had appeared by the side of normal forms; by a much lighter colour they showed a disposition to lose their colour entirely. The variability of the different rose variants is not the same; the form, obtained by the concentration

experiment (3) produces, more readily than the rose variant (2), as well red normal forms (1) as white ones (4). For the rest, this more variable variant has also proved to remain constant when quickly transplanted.

Cases of atavism are frequently observed in these experiments. Thus, for example, the production of the normal form from *viscosus* (6) may easily be seen if the latter grows for a fortnight without transport on a bouillonagarplate; along the margin of the streaks some few normal colonies (1) will then become perceptible.

The *albus*-variants, also have a disposition to throw off a few red normal forms, but they do so only after growing for weeks or months on bouillon-agar; at first they are very constant.

The to a certain extent completely regular production of the same variants of *Bacillus prodigiosus*, suggests the existence of variability in a special and determined direction, of orthogenesis, as EIMER expressed it.

As under different nutritive conditions the same variant may appear, the food itself cannot be the stimulus; there must be, as said above, another cause in the interior of the cells, which, for *B. prodigiosus*, seems only active in an alkaline environment.

On the other hand, the food, in a wider sense, has certainly a decisive influence on the variability, albeit indirectly. So we considered already the influence of the alkaline reaction of the medium if this alkali is produced by the microbes themselves. Another example is the following. As well in malt-wort as in bouillon the *viscosus* variant is regularly produced; but from malt-wort the *auratus* variant, which so readily takes rise in bouillon, is not obtained at all. Indeed, every culture condition gives a peculiar but constantly returning mixture of variants, differing both quantitatively and qualitatively from that found under any other conditions. But the real factors here active could not as yet be detected.

From the foregoing the following results may be derived.

1. *Bacillus prodigiosus* produces as well qualitative, as gain- and loss-variants, all obtained with certainty by determined experiments; the stock-form is always found unchanged in the same culture with the variants.

All the variants are from their origin as constant as their stock. The true factors which govern the variability in these experiments are still unknown.

2. By rapidly repeated re-inoculations and by other methods, nor-

mal form and variants may be kept constant, as it seems for an unlimited length of time.

3. All the variants vary in a way analogous to that of the normal form, thus, the *auratus*-variant produces an *auratus*-slimevariant, which must be considered as a gain-variant, and an *albus*-variant, which must be taken for a loss-variant.

The natural variety *B. Kieliensis*, which approaches the *auratus*-variant, also varies in an analogous way. The variation thus seems to be directed or orthogenetic.

4. Gain-atavism in loss-variants and loss-atavism in gain-variants, can be obtained with certainty by determined experiments. Qualitative variants, too, may give rise to atavism.

5. The experimental variants of *B. prodigiosus* have not yet been found in nature. From another bacterium, *Bacillus herbicola*, a variant, took rise which I had before repeatedly isolated from nature and which I had taken for quite another species.

6. The variants of *prodigiosus*, and this holds good for many other microbes also, differ from each other and from their stock forms in the same way as closely related natural species or varieties do among each other. But their disposition to atavism is much more pronounced.

7. The sub-variants, e. g. the rose variants of different colour-intensity, arise in the same way as the chief variants and possess the same degree of constancy.

Physics. — "*Researches on magnetization at very low temperatures.*" By PIERRE WEISS and H. KAMERLINGH ONNES. Communication N°. 114 from the Physical Laboratory at Leiden.

§ 1. *Object of the research; results.*

a. Introduction. The extension of LANGEVIN's [1]) kinetic theory of magnetism to all ferromagnetic phenomena by means of the hypothesis of the molecular field [2]) rendered the testing of deductions from this hypothesis by experimental data of great importance. The first results of this comparison were very encouraging; in some respects a remarkable correspondence was found. For instance the curves

[1]) LANGEVIN. Ann. Chim. et Phys. 8 Sér. t. 5, p. 70; 1905.
[2]) P. WEISS. Journ. de Physique 4e Sér. t. VI, p. 661; 1907.

calculated for the intensity of the magnetization at saturation as a function of the temperature corresponded very well with those which had been found experimentally for magnetite at temperatures above the ordinary. Moreover, the law determining the susceptibility above the CURIE-*point* [1]) developed from the hypothesis of the molecular field was found in CURIE's experiments, and in others which will soon be published, to be accurate over a temperature range of some hundreds of degrees. Finally the sudden changes in the specific heat at the CURIE-point were in correspondence with the values calculated from magnetic data. But other observations do not correspond so well with the theory. Fig 1, Pl. I in which the theoretical curve for the change of saturation-magnetization with temperature is shown by the full curve *a*, also shows the experimental results for magnetite, and the corresponding curve, *b*, for nickel [2]). The last curve is drawn to such a scale that the best possible correspondence with the theoretical is obtained at the CURIE-point. In contrast with what was found for magnetite, nickel shows a deviation from the theoretical gradually increasing over the whole curve. Iron and cobalt behave practically the same as nickel. When all this is taken into consideration it is seen that the hypothesis of the molecular field is of the nature of a working hypothesis; the partial confirmation shows that the hypothesis contains a kernel of truth, and from the experimental deviations one will have to see how it should be modified or extended while still retaining its essential features.

It is not probable that these modifications will attack the property of reacting against the orientation by the magnetic field that has been ascribed to the kinetic energy, or that they will come into conflict with the manner in which the MAXWELL-BOLTZMANN partition law has been employed. Not only are these hypotheses of fundamental import, but they are still further forced upon our consideration by the ease with which they account for the fact that for paramagnetic substances the susceptibility varies inversely as the absolute temperature — an experimental law that is one of the most firmly established for a number of substances. In their important investigations upon the magnetization of the elements, of which an account was given at the last

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[1]) In this Communication we shall give the name CURIE-point to the temperature at which spontaneous ferromagnetism ceases. This is by no means inconsistent with CURIE's idea that the transformation temperature is a function of the strength of the field, since the temperature at which *spontaneous* ferromagnetism ceases is the temperature obtained by reducing the field to zero.

[2]) According to preliminary measurements. Accurate experiments upon the three metals and magnetite are in progress.

meeting [1]), H. DU BOIS and HONDA have, it is true, shown that this law of the dependence of susceptibility upon temperature is not generally valid, and that paramagnetism also occurs which is independent of the temperature or increases with increasing temperature. But it is by no means the case that the foregoing hypotheses should be discarded on that account; what we learn from experiment in this case is only that these suppositions are not sufficient to explain magnetism as a whole. In particular it will be necessary to revise LANGEVIN's hypothesis that the magnetic moment of a molecule is constant, or at least quasi-constant, and also that concerning the nature of the mutual action of the molecules, which until now has been represented by the introduction of the molecular field. For an at low estimation of the value of both of these hypotheses, experiments temperatures are especially valuable.

For it is only at the absolute zero that the magnetization gives the sum of the molecular magnetic moments, as it is only then that heat-motion can no longer prevent the magnetization from attaining its full value; and at low temperatures, too, is the strongest demonstration of the mutual action of the molecules to be expected, since they are then at the smallest possible distance from each other.

b. Ferromagnetic substances. We have, therefore, aimed at the continuation of the curves connecting magnetization and temperature in the three ferromagnetic substances and in magnetite down to the neighbourhood of the absolute zero. By utilising the methods and appliances [2]) suitable for long-continued accurate measurements at such constant temperatures as are obtainable with liquid hydrogen, we have been able in our measurements to reach a temperature of 20°,3 K. with hydrogen boiling under atmosphere pressure, and of 14°,0 K. with hydrogen near its melting point. The number of degrees on the absolute scale which separate these experimental temperatures from the absolute zero is but such a small fraction of the number between the absolute zero and the CURIE-point (even in the case of nickel this number is still so much as 648 Kelvin degrees) that, considering the nature of the curves, we may regard the saturation-magnetization at the absolute zero as being determined by our experiments. All this, of course, with the proviso that the phenomenon in the region to which extrapolation is carried should give no occasion for adopting another point of view. Since the object of the measurements was a determination of the saturation-magnetization, it seemed suitable to direct the expe-

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[1]) These Proceedings Jan. 1910.

[2]) H. KAMERLINGH ONNES, these Proceedings Sept. '06, Comm. Leyden N°. 94*f*.

riments towards obtaining data for magnetization in strong fields, and from these the deduction of the law according to which the magnetization approaches its limiting value. But the method chosen for the magnetic measurements, viz: the determination of the maximum value of the couple exerted by a magnetic field of varying direction upon an ellipsoid of the experimental substance, was, as we shall presently show, less suitable for this determination of the law of approach than for comparisons of the magnetizations of the substance in the same field at different temperatures. The data to determine the law of approach were therefore made the subject of a separate investigation [1]) This gave the following values for the difference between the magnetization in a field of 10000 gauss and that in the limiting case:

Iron	0.08 %
Nickel	0.1 „
Cobalt (soft)	1.1 „
Magnetite	0.19 „

For these substances, the cobalt excepted, the approach of magnetization as a function of the strength of the field is hyperbolic, so that in a field of 20000 gauss, which we reached in our present experiments, the above differences were reduced to half their values. Observations by the ellipsoid method in different fields and at both low and ordinary temperatures have not, indeed, enabled us to test the law of approach, but they show sufficiently well that there is no essential difference between the behaviour in this respect at the two temperatures; and that at low temperatures, as could have been supposed the magnetic hardness does not assume an excessive value, the molecules hindering each other in assuming a new direction.

Further, by means of comparative measurements, magnetizations at ordinary and at low temperatures in fields of great strength were compared, and it was found that the ratio between the two is pretty well independent of the strength of the field. Thus, leaving the result uncorrected for the dilatation between the two temperatures (see note 2 on p. 11) we found for the ratio of the intensity of magnetization at 20°,2 K. and at ordinary temperature the following:

Nickel	(17°.3 C.)	1.0548
Iron	(20° C.)	1.0210
Magnetite	(15°.5 C.)	1.0569

The exact value of the ordinary temperature is given between brackets. In § 5 it will be explained why the experiments with

[1]) P. Weiss, Arch. des Sc. phys. et nat. février 1910 and Journ. de Phys. 4e Sér. t IX. avril 1910.

cobalt have not been brought to a conclusion. It is difficult to know exactly the degree of accuracy of these results. Experimental work in every branch was carried out so that an accuracy of 1 in 1000 or even higher could be expected. But when one considers the disturbing influences which made themselves felt in the experiments upon cobalt, it seems rather incautious — and this is particularly the case with the magnetite measurements — to ascribe to the results an accuracy greater than 0.5 $^0/_0$, even though the occurrences which have thrown suspicion upon the cobalt measurements were nearly absent in the case of the other substances, and though in all its properties and particularly in its extraordinarily large magnetic hardness cobalt stands evidently alone. Since our experiments indicate these causes of uncertainty, they show how a higher degree of accuracy may be reached if so desired. The present accuracy is quite sufficient for the treatment of various problems.

The experiments with iron and magnetite were carried to 14°,0 K. The change of magnetization between 20° K. and 14° K. is too small to be expressed in figures. These experiments, therefore, only extend down to 14° K. the temperature region within which the diminution of the kinetic energy and the approach of the molecules to each other do not occasion the appearance of a single new phenomenon.

The portions of the curves for nickel and magnetite which have been newly obtained are given by broken lines in fig. 1, Plate I.

Magnetite is of particular importance on account of the perfect correspondence between observation and theory over the greatest portion of the region between the CURIE-point and the absolute zero, and on account of the occurrence of a deviation of observation from theory only at low temperatures. Here, theory gives for the ratio between the magnetizations the value 1.139 instead of the value given above, 1.057. The result that theory and experiment clearly differ at these temperatures is corroborated by earlier experiments upon four samples of different kinds of magnetite, two obtained from natural crystals, the third from a fused natural crystal, and the fourth from artificial magnetite. These gave the following values for the ratio between the magnetizations at the temperature of solid carbon dioxide (— 79° C.) and ordinary temperature:

1.033	ordinary temperature	16° C.
1.042		23°.2
1.043		24°
1.037		21°.5
mean 1.039		21°.2 C.

while theory gives 1.053 for the same temperature.

An analogy thus seems to exist between this and compression and expansion by heat, for which VAN DER WAALS's theory and law of corresponding states are supported better as a rule in the neighbourhood of the critical point than at low reduced temperatures where the ideal representations of the molecule and of molecular attraction no longer cover the phenomena sufficiently well and the differences between the specific properties of the real molecules appear.

The hypothesis that molecular magnets are essentially invariable would be established conclusively if there existed simple relations between the magnetic moments as calculated per atom, which one might be led to suspect from the increase by regular steps of the saturation-magnetization of the three metals.

The following table in which the numbers in the first column are taken from the paper [1]) referred to above and in which the relative increase for cobalt is estimated from comparison with iron and nickel shows that this is not the case. The data are not corrected for the dilation (see note 2 on p. 11).

	Specific saturation at temp. ( ).	Increase by reduction to low temp.	Specific saturation at low temp.	Atomic weight or $^1/_3$ mol. wt.	Moment of gram-atom.
Ni	54.6 (17° C.)	1.0548	57.6	58 7	3381
Co	162 (17° C.)	1.01	163.6	59	9650
Fe	217 (20° C.)	1 0201	221.6	56	12410
$FeO^1/_3$	90.75 (15°.8 C.)	1.057	95.9	77.33	7417

In connection with this we must not lose sight of the fact that although the proof that the above magnitude is of great significance may have escaped us, still there is nothing whatever to justify an opposite conclusion.

When we look upon our measurements as a whole we remain inclined to retain the hypothesis that in ferromagnetic substances the magnetic atom does not in itself change much with temperature. There were indeed reasons for questioning if this approximate invariability, granting that it was proved in other circumstances, still existed at extremely low temperatures. Electrical resistance of metals, phosphorescence of sulphur compounds, absorption of light by the salts of the rare earths with or without magnetic field, all, at very low temperatures, exhibit characteristics that one may

[1]) P. WEISS. Arch. des Sc. phys. et nat. and Journ. de Phys. 1910.

try to explain by ascribing them to forces exerted by ponderable matter upon electrons; these forces in that explanation become of primary importance when the temperature sinks to that of liquid hydrogen, and it is ascribed to them in particular, that they make the current-carrying electrons in metals suffer an important diminution in number at very low temperatures by their being, as it were, frozen to the atom by the low temperature [1]).

It would also be possible that the motions of the electrons which cause magnetism while remaining constant or changing not much at other temperatures, begin to show considerable changes at very low temperatures.

The negative result that nothing happens even at the lowest temperatures, which should throw doubt upon the relative smallness of the variability of the magnetic atom itself, is not perhaps without importance when regarded as a means of weighing the value of the above assumptions regarding the phenomena mentioned, or as means of separating the group of electrons which occasion magnetism from groups which form the prime factors of other phenomena.

c. Vanadium, chromium, manganese. The question has often been asked if a gap which cannot be bridged over exists between the ferromagnetic metals of the iron group on the one hand and the paramagnetic metals of the same group on the other, or that the latter metals should also exhibit a very low CURIE-point if the temperature were sufficiently lowered.

CH. ED. GUILLAUME [2]) says with reference to the HEUSLER alloys of Mn, Al, Cu and Mn, Sn, Cu which are ferromagnetic: "The reason for this can be found in the fact that aluminium or tin when compounded with manganese, a metal from the magnetic group, raises its transformation temperatures, which, following an hypothesis already suggested by FARADAY, ought to lie very low." It can indeed be seen that aluminium and tin raise the melting points of various alloys which they form with other metals (the series Al—Au, Al—Sb, Na—Sn) and seem to possess the general property of raising temperatures of transformation.

We might, therefore, expect that vanadium, chromium, and manganese should at very low temperatures exhibit either the characteristics of ferromagnetism (magnetization not proportional to strength of field,

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[1]) Cf. H. KAMERLINGH ONNES, Comm. fr. the Leyden labor. Suppl. n°. 9, p. 27 1904 and P. LENARD, H. KAMERLINGH ONNES and W. E. PAULI, These Proceedings June 1909, Comm. fr. the Leyden Laborat. n°. 111, p. 3, note 2 1909.

[2]) Ch. ED. GUILLAUME. Actes de la Soc. helv. der Sc. nat. Vol. I p. 88. 1907.

saturation, hysteresis) or, in conformity with CURIE's law, a strongly increased paramagnetism. The susceptibility at the temperature of solid hydrogen should be about twenty times as great as at ordinary temperature [1]). At this time we were not yet aware of the results published last month by H. DU BOIS and HONDA [2]), from which it appears that the inverse proportionality of paramagnetism to the absolute temperature is but one of the possible cases. To get an idea of the order of magnitude of the expected phenomena we may suppose that the paramagnetic γ iron still exists at 14° K. with the same CURIE constant (product of absolute temperature by susceptibility). In that case a value of about 400 is found for the magnetization of this substance in a field of 20000 Gauss.

Some time ago GEBHARDT [3]) determined the susceptibility of manganese at ordinary temperature and found $K = 322.10^{-6}$ (density 6.4). The above calculation gives a value 134 for the magnetization of this substance in the same circumstances. And as the deflection in our apparatus is proportional to the square of the magnetization, one would obtain a deflection smaller in the proportion of 18 in the case of γ iron or 160 in the case of manganese than that which was found for iron at the ordinary temperature; as this was 100 cm. the manganese deflection should still be quite easily readable.

When we now introduced into our apparatus roughly formed ellipsoids of MOISSAN vanadium and GOLDSCHMIDT chromium and manganese in succession, the awaited change did not appear. In every case the deflection at the temperature of solid hydrogen as well as at that of hydrogen boiling under atmospheric pressure remained the same as it was at ordinary temperature, that is, to a few tenths of a millimetre, and these must be ascribed to the magnetism of the suspending apparatus. There was therefore no ferromagnetism and we were obliged to choose between the following two hypotheses for these substances. We were either dealing with paramagnetism of a new kind or with diamagnetism, which is also

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1) A similar supposition formed the starting point of a research by H. KAMERLINGH ONNES and A. PERRIER, which will shortly be published, and is closely connected with the present research. This investigation has been taken to hand at the same time with the present subject. Using the method of the maximum couple and the hydrostatic rise the magnetizations of liquid oxygen at various temperatures and of solid oxygen at the temperatures of boiling and solidifying hydrogen were measured. The increase of the magnetization at low temperatures was found to be very great, though not so much as was expected, and a distinct deviation from CURIE's law and a characteristic curve were found.

2) H. DU BOIS and HONDA l. cit.

3) GEBHARDT. Inaug. Dissert. Marburg 1909.

found in copper while most of the salts of this metal are paramagnetic DU BOIS and HONDA's paper in which these three metals are classified under those whose paramagnetism is invariable or increases with the temperature shows that the first assumption is the correct one. The behaviour of copper with the present research made us consider the other hypothesis a reasonable one.

One could always assume that the paramagnetism, which, as a general rule is ascribed to the metallic manganese, results from the presence of its oxides, which are strongly magnetic, or of a small quantity of iron. To put this assumption to the proof we prepared very pure manganese from MERCK's pure chloride, which had been proved to be free from iron. The preparation was accomplished by electrolysing the salt between a cathode of distilled mercury and an anode of iridium alloyed with 40% of rhodium which is not attacked by the chloridion. The almagam obtained in this way was separated in a stream of pure, dry hydrogen. In this way a grey powder was obtained which when compressed in a glass tube as a mould took the shape of a solid rod. A rod prepared in this manner exhibited paramagnetism. A glass tube with the powdered manganese was also paramagnetic. The same manganese contained in a magnesia boat was thereupon fused in an electric resistance furnace and in an atmosphere of hydrogen. In this way an ingot was obtained which was covered with a light oxidised crust. It was found impossible to grind away this crust with quartzpowder, since the metal was of the same hardness as quartz. Emery could not be used as it is magnetic. The impure crust was therefore turned off with a diamond tool, and a small cylinder of pure substance was obtained.

This cylinder was found to be *ferromagnetic*. Fig. 2 Pl. I gives the hysteresis curve for this substance. The maximum value of the specific magnetization is 100 times weaker than that of iron, and the coercive field is 670 gauss, that is to say, 10 times as strong as the coercive field of steel which is used for the preparation of good permanent magnets. This peculiar substance seems moreover to have striking magneto-crystalline properties. The rod was strongly attracted between the poles of a magnet and placed itself perpendicular to the field.

Manganese of the same degree of purity can therefore occur in two states: paramagnetic and ferromagnetic. GEBHARDT's experiments give a susceptibility five times greater than that observed by DU BOIS. If GEBHARDT's powder was not impure or oxidised, it is thus possible that there are two paramagnetic states.

[1]) SECKELSON, Wied. Ann. LXVII, p. 37, 1899.

As regards the ferromagnetism of manganese, this had already been observed by SECKELSON [1]) with electrolytic manganese which was liberated at 100° C. from the chloride upon a platinum wire, and with a regulus prepared by BUNSEN from manganese fluoride. The very indefinite observations concerning the magnetization which he published do not contradict our measurements.

By a more direct method we have proved the absence of strong magnetism in vanadium, chromium and manganese at low temperatures. For this purpose we introduced ellipsoids of the three substances into a narrow unsilvered vacuum tube whose walls were separated by the smallest possible distance; this was placed in a second similar tube also as narrow as possible and filled with liquid air. We then determined the distance from the poles such that the ellipsoids were attracted from the bottom of the tube to the poles of the magnet. This experiment was made first with the inner tube empty, and then with the inner tube filled with liquid hydrogen. The following results were obtained:

	Ordinary temperature	In liquid hydrogen
Vanadium	Not attracted	The same as at ordinary temperature
Manganese	Attracted from distance of 6 to 8 mm.	
Chromium *a*	" " " " 12 "	
Chromium *b*	" " " " 60 "	

The results for Chromium *b* which probably contained a small splinter of iron must be rejected. We also found further that a crystal of iron sulphate at ordinary temperature was attracted from a distance of 25 mm. while in liquid hydrogen it was attracted almost from the base of the magnet. Thus the weak magnetization of the three metals was found to be practically invariable, while the iron sulphate exhibited a very great increase in magnetic properties.

This experiment is well adapted for displaying the characteristic difference between the two groups of substances and is a typical example of the significance which even the simplest experiments acquire within the fallow region of very low temperatures.

§ 2. *Methods and apparatus.*

a. Discussion of the method of the maximum couple. We measured the intensity of magnetization by measuring the couple exerted on a prolate ellipsoid of revolution of the experimental substance arranged so that the angle of the field with the major axis of the ellipsoid might be varied. The expression for the couple is

$$M = (N_1 - N_2)\, I^2 v \sin \varphi \cos \varphi$$

where N_1 and N_2 are the coefficients of demagnetization of the ellipsoid, I, the intensity of magnetization of the substance, v the volume, and φ the angle between I and the major axis of the ellipsoid. The maximum value of this couple is

$$M = \frac{N_1 - N_2}{2} \cdot I^2 v$$

for $\varphi = 45°$. Hence to measure I it is not necessary to know either the strength or azimuth of the field which yields the maximum couple. To make use of these methods the ellipsoid is suspended from a torsion-spring whose displacement is determined by a mirror-method, and an electromagnet turning round a vertical axis is used. The method has already been described [1]). Its advantages consist of the small range over which strong fields are necessary and the extreme simplicity of the relative measurements.[2]) We shall now discuss two sources of error which affect it and which, although they may be made as small as one wishes in theory, render it less suitable for the search after the law of approach to saturation, although they do not take away from its value as a method of comparing in the same field two successive and slightly differing states of the same substance.

Influence of inhomogeneity of the field.

The ellipsoid is placed in the centre of a magnetic field possessing the symmetry of a body of revolution. The strength of the field at this centre is a maximum for a displacement in the plane of the equator, y, and a minimum for a displacement in the direction of the x axis. It it given by the series

$$H = H_0 + \frac{x^2}{1 \cdot 2}\left(\frac{\partial^2 H}{\partial x^2}\right)_0 + \frac{y^2}{1 \cdot 2}\left(\frac{\partial^2 H}{\partial y^2}\right) + \ldots,$$

which, remembering the equation $\Delta V = 0$ for the magnetic potential V, and converting to polar coordinates r and θ, transforms into

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[1]) P. Weiss, Journ. de Phys. 4 sèr. t. VI, p. 655, 1907.

[2]) For comparing the intensities of magnetization I and I' at two temperatures we have to take into account that $v = m/d$, m being the mass of ellipsoid and d its density, so $\frac{I}{I'} = \frac{\sqrt{M}}{\sqrt{M'}} \cdot \frac{\sqrt{d}}{\sqrt{d'}}$ and $\frac{\sigma}{\sigma'} = \frac{\sqrt{M}}{\sqrt{M'}} \cdot \frac{\sqrt{d'}}{\sqrt{d}}$. The dilatation at the low temperatures and therefore the proportion of d and d' being unknown, we have omitted the correction for the difference of this proportion and unity, the value of which may be estimated at 0,004. [Added in Translation].

$$H = H_0 + \frac{r^2}{2}\left(\frac{\partial^2 H}{\partial x^2}\right)_0 \left(cos^2\,\theta - \frac{1}{2}\,sin^2\,\theta\right).$$

Now, the energy of a volume-element dv of the ellipsoid which we consider to be very long and magnetized with equal intensity I in the direction of the field, is

$$W = -IHdv$$

and therefore the moment of the couple exerted by the field on this element is

$$dM' = \frac{\partial W}{\partial \theta} = \frac{3}{2}\,dv\ .\ I\left(\frac{\partial^2 H}{\partial x^2}\right)_0 r^2\,sin\,\theta\,cos\,\theta.$$

The couple exerted by the field H_0 on the ellipsoid is

$$M = (N_1 - N_2)\,I^2 v\,sin\,\varphi\,cos\,\varphi.$$

In very strong fields the condition is fulfilled that the magnetization is parallel to the external field nothwithstanding the demagnetizing forces of the ellipsoid, and therefore $\theta = \varphi$.

Hence the disturbing moment dM' varies with azimuth of the substance in exactly the same manner as the chief couple M. The maximum value of the couple dM' is

$$dM' = \frac{3}{2}\,dv\,I\,.\left(\frac{\partial^2 H}{\partial x^2}\right)_0 r^2,$$

which for the whole ellipsoid gives

$$M' = \frac{3}{10}\,I\left(\frac{\partial^2 H}{\partial x^2}\right)_0 v\,.\,a^2,$$

where a is the semi-major-axis of the ellipsoid.

Let us now make the assumption that the field changes conformally, and let us call the field 1 cm. from the axis in the direction of the y-axis $(1 - \varepsilon)\,H_0$, then

$$\varepsilon H_0 = \frac{1}{4}\left(\frac{\partial^2 H}{dx^2}\right)$$

and therefore

$$M' = \frac{6}{5}\,I\varepsilon\,H_0\,v\,.\,a^2$$

and the ratio between the maximum values of the couples is

$$\frac{M'}{M} = \frac{6}{5}\,\varepsilon\,a^2\,\frac{H_0}{(N_1 - N_2)\,I}.$$

With constant magnetization, therefore, the second last equation shows that the disturbing couple increases proportionally to the strength of the field. In the IH diagram, a sloping instead of a

horizontal asymptote will be found. This was shown clearly in some of the foregoing experiments. If the field is constant the disturbing couple increases with I. Therefore, if in the measurements with the greatest values of I for which the experiments are carried out, made with a certain apparatus the disturbing couple does not make its presence felt, then *a fortiori* is it negligible for the smaller values of I. The last equation shows that the *relative* value of the couple for non-uniformity of the field increases as the intensity diminishes. Hence it is to be feared particularly when one works with small magnetizations, and when, to increase the sensitivity of the apparatus, the torsion spring is replaced by a weaker one.

For the purposes of our measurements it is sufficient to get an idea of the order of magnitude of the error. For this purpose the non-uniformity of the field was measured for three different values of y; it was found to be proportional to y^2 with $\varepsilon = 0.0087$ as factor.

With $a = 0.15$ it follows that $\frac{M'}{M} = 0.00023 \frac{H_0}{(N_1 - N_2) I}$. For the ellipsoids used $N_1 = 1.90$ and $N_2 = 5.59$, and $(N_1 - N_2) I$ is almost 6600 gauss for iron and 1800 gauss for nickel. Hence, for iron the correction is scarcely 1 in 1000, while for nickel it increases to some thousandths.

Reaction of the ellipsoid on the pole-pieces.

When the ends of the ellipsoid come into the immediate neighbourhood of the end surfaces of the poles, they exert a noticeable influence upon the distribution of magnetism in the pole-pieces, and the couple becomes increased thereby. This fact was established by previous experiments with a larger electromagnet with flat pole-pieces of 15 cm. diameter. In these experiments was measured the couple exerted upon an ellipsoid with various distances between the poles by a field of the constant value of 9770 gauss regulated each time by passing the required current. In this way the following values were obtained for an iron ellipsoid 9 mm. long and 4 mm. thick.

Distance between poles	Maximum couple
9 mm.	335.6
15 „	320.45
23 „	319.32
35 „	319.18
47 „	319.08

The law according to which this magnitude changes shows that the change is not a consequence of the non-uniformity of the field; for just when the disturbance reaches its greatest value, the field is most regular owing to the closer approach of the flat pole-pieces. For distances of 23 mm. and greater the influence is insignificant, and the couple is constant.

b. Electromagnet. From what has been said about the influence of the ellipsoid and the pole-surfaces it follows that the distance between the poles should be about three times the length of the ellipsoid. The total thickness of the four walls of the DEWAR tubes and of the holder (§ 2*c*) could not be made smaller than 5 mm. Hence, keeping account of the difficulty on the one hand of obtaining strong fields of wide extension and on the other hand of reducing very small ellipsoids to the correct form, we decided upon an inter-pole distance of 9 mm. and a length of 3 mm. for the ellipsoids.

With this distance comparatively strong fields (up to 25000 gauss) may be excited with a magnet whose cores are 9 cm. in diameter. The electromagnet of this power which was used in these experiments has already served for magnetic experiments at high temperatures. It has already been described [1]) and is represented diagrammatically in fig. 1 Pl. II. Comparatively light (132 KG.) and, taking its power into account, easily transported, it was possible to study it in Zurich and to use it in Leyden. Hand-wheels, whose position is read from divided circles, communicate a horizontal micrometric movement to the pole-pieces.

The magnet turns upon a vertical axis and for that purpose is mounted upon a ball-bearing support. The azimuth is determined by means of a fixed mark on a cylindrical scale E_2 attached to the movable portion of the supporting base. Each of the coils has 1500 turns of 2.5 mm. wire and has a resistance of about 2 ohms. As the coils are arranged for a current of 10 amp. under ordinary circumstances, and as the current can for a short time be increased to 25 amp. the number of ampere-turns at one's disposal may reach as high as 75.000. The water circulation E_5 between the double walls of the coils has this immediate advantage that the duration of an experiment may be doubled, but it is chiefly of importance in protecting the pole-pieces from heat. Such a heating would lead to various difficulties, of which one of the worst would be that the strength of the field would noticeably change, for the expansion of the com-

[1]) G. ZINDEL. Revue électrique 20 Juin 1909 and Elektrot. Zeitschr. XXX, p. 446, 1909.

paratively long core by heat could distinctly alter the comparatively short distance between the poles.

c. Cryogenic apparatus. As it was necessary to shut off from the air the space in which the ellipsoid was freely suspended since it contained liquid hydrogen and its vapour, a fairly complicated cryogenic apparatus had to be employed. This is shown diagrammatically in Pl. II fig. 1 and in section in fig. 3. The apparatus consists chiefly of three tube-shaped portions which, naming from outside inwards, we call the *cover*, the *adjusting tube* f, and the *holder* b. The cover consists of a silvered vacuum tube A, a brass tube B, a glass tube C, and a cap D which shuts off the apparatus from the air.

Holder. The ellipsoid a (figs. 3 and 5) can turn round a vertical axis with the holder b in which it is fixed. For the greater part of its length the holder is made from a tube b_2 of german silver — a substance that is rigid, little magnetic, and a bad heat-conductor. The lower end is joined to a copper rod b_1, which has only a very weak inherent magnetism. The holder is connected to the rod k by the spiral spring g_1 (g_1 was used for iron and cobalt; the weaker spring g_2, which was used for nickel and magnetite is shown at the side). To make the equilibrium stable and to prevent the ellipsoid from being attracted to the poles of the magnet the holder is held fast underneath by a wire of platinum-iridium of 0.1 mm. diameter, for the torsion of which a correction need hardly be applied (§ 4).

The tube b_2 and the rod b_1 are carefully adjusted on the lathe, and the ellipsoid a (fig. 4) is fixed carefully in a cylindrical opening, the diameter of which is equal to the minor axis of the ellipsoid. If the ellipsoid is nickel or magnetite it can be fixed in position with a little wax. With iron and cobalt, however, the ellipsoid is subject to such strong forces that it is necessary to clamp it fast by covering it with a thin piece of sheet copper and then driving it forcibly into the opening. The turning of the ellipsoid is transmitted through the rod b_1, and the thin-walled german-silver tube [1]) b_2 to the mirror h. From the mirror through the opening f_{10} and the window C_2 (figs. 1 and 3) the torsion of the spring g_1 is read. A

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[1]) A slight twisting of this tube is of no account. Only that portion of the apparatus between the mirror and the cap acts as a spring. Twisting of the portion of the apparatus below the mirror only transmits the couple to that spring, its sole effect is to slightly, but not noticeably, alter the azimuth of the magnet.

glass scale 1.5 meters long, and subdivided into half millimeters is used; it is placed at a distance of 4.325 m. and is illuminated by spherical mirror strips [1]). The tension of the spring is regulated by the rod k (fig. 3), which passes through a stuffing box D_2 in the cap D. Vertical motion is communicated to k by turning the nut D_4 and at the same time preventing the motion of D_5. The tension is read through the opening f_{21} from the pointer l on the scale b_{20}. Before mounting the apparatus, that division of the scale b_{20} is determined which corresponds with the tension that is to be used, by suspending known weights from the stretching wire.

The apparatus is, like a stretched string, very liable to start vibrating under the influence of small impulses. This tendency is counteracted by immersing the vanes of a vane-damper b_3 (fig. 6 and fig. 3) in oil contained in a circular vessel divided into different chambers by the partitions b_{34}. These partitions are attached to a cylinder which turns with slight friction in the adjusting tube and is therefore carried round by the vanes b_{30} whenever the holder must experience a somewhat greater torsion (§ 4) [2]). The vanes must be wholly immersed in the oil so as to ensure that capillary reactions do not bring forces into play (see § 4), whose torsional effect could not be neglected. In strong fields the torsion oscillations are damped extremely well by the FOUCAULT currents.

The whole holder and spring hang in the adjusting tube f the upper end of which is screwed to the cap D; this cap also carries the rod k, and is itself supported by the glass tube C. The adjusting tube, consisting of the portions f_2, f_3, f_4, f_5, is three times diminished in cross-section. The lowest portion f_5 is narrow and surrounds the rod b_1 of the holder as closely as possible. Against the bottom f_6 (fig. 5) rests the cone c, which is soldered to the wire d and serves to keep it taught. A slit in the bottom allows the conical portion to be placed in position (fig. 5). When the apparatus is put together the adjusting tube sinks into the DEWAR vessel A so that the thin tube f_5 is centred in the narrow portion of the vacuum tube. The adjusting tube as well as the tube b_2 of the holder is made of german-silver.

To mount the adjusting tube already containing the holder in the cover, the cap D is screwed to a bronze ring cemented to the glass tube C of the cover; the screws D_7 are tightened, and the junction

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[1]) H.KAMERLINGH ONNES, Comm. fr. the phys. Lab. Leiden, n°. 25. (1896).

[2]) It is essential to free the oil beforehand from volatile substances, and also to prevent the accumulation of air bubbles under the oil, since the apparatus has to be completely evacuated after it is put together.

is made air-tight by means of the rubber sleeve D_6 which is smeared with rubber solution and bound with copper wire. The lower end of the glass tube C is cemented to a second bronze ring, which is soldered to the brass tube B of the cover. To the centre of this brass tube is attached a ring B_3 carrying the bolts of the supporting rods B_4 which hold the vacuum glass in position.

The DEWAR tube itself consists of a narrow lower portion A_2 completely silvered and a wider upper portion that is silvered up to A_3 (the upper portion is left transparent so that we might be sure that we were not allowing too much liquid hydrogen to enter the glass). It fits into the brass tube B_1 and is protected by a wooden ring. The supporting rods B_4 keep the vacuum tube in position and at such a height that it is just clear of the wooden safety ring. Fig. 7 shows how, by means of the screw B_{50}, the vacuum glass protected by a layer of paper is clamped to the thin brass ring B_5, to which are attached the ends of the supporting rods B_4. The lower portion of the vacuum tube has an external diameter of 8 mm. and an internal diameter of 5 mm. The glass walls are 0,5 mm. thick, which leaves only 0,5 mm. as the distance between the two silvered walls.

The apparatus is centred by placing it on an auxiliary support by means of the ring B_2. Before the vacuum tube it yet in position, the narrow portion f_5 of the adjusting tube is adjusted by a central ring in an adjustable centring-plate. The loose ring is then removed from the plate and a second is fitted such that it just fits the narrow portion of the lower end of the vacuum tube. The nuts B_{31} serve to bring the vacuum glass to its proper position, and, as before, it is made air-tight by a rubber sleeve B_6, which is smeared with rubber solution and bound with copper wire. By adopting this method of attaching the vacuum tube one need not fear alteration of the cover when the apparatus is evacuated, and only small further adjustments are necessary for recentring the apparatus after evacuation.

In the tube B is soldered the steel capillary θ_3 (figs. 1 and 3) of a helium thermometer [1]) with german silver reservoir θ_1 (figs. 3 and 7) and glass stem θ_4, which is permanently attached to this portion of the cover. The quantity of helium is so chosen that at the boiling point of oxygen the mercury stands at a mark in the lower portion of the stem, and at the melting point of hydrogen at one in the upper portion. If, as is the case with hydrogen boiling under ordinary atmospheric pressure, the temperature is sufficiently well known

[1]) Compare the apparatus for the liquefaction of helium. H. KAMERLINGH ONNES These Proc May/June 1908, Comm. Leid. N°. 108.

without reading the thermometer, the thermometer is still necessary, however, to indicate the position of the upper surface of the liquid gas which is no longer visible beneath A_3. As soon as the level sinks below the upper end of the reservoir θ_1 of the thermometer, the mercury in the stem θ_4 sinks.

d. First the electromagnet is adjusted which operation is independent of the centring of the adjusting tube, the holder and the vacuum tube. The axis round which it turns is made vertical, and then the pole distance is centred round this axis. Next the centre of the truncated spherical socket G_{10} (fig. 1, 2 and 3) is made to coincide with the axis round which the magnet turns. It is supported by a plate which is attached by two beams to the freestone pillar ℭ. The cryogenic apparatus is then brought from its auxiliary support and arranged in its proper position by placing the ball-shaped portion of the surface of the ring B_2 in the concentric socket G_{10}; the centring of the narrow portion of the vacuum tube on the turning-axis of the magnet is completed by means of wing nuts on the ring B_2. This centring must be done with great accuracy, for the magnet must turn freely and the distance between the vacuum tube and either pole is not more than half a millimetre. It can, however, easily be accomplished to 0.25 mm.

e. Liquid hydrogen is introduced into the apparatus by a german silver tube B_7 (cf. Comm. N°. 94*f*). The gas formed by evaporation escapes through B_8 (figs. 3 and 1) and through the valves K_1, K_2 (fig. 1) to a gasometer or to a vacuum pump. By means of the valves the vapour pressure is regulated, and its value is read on a manometer H which at the same time acts as a safety valve. In experiments made in the neighbourhood of the melting point of hydrogen the pressure was kept slightly above that of the triple point.

Before introducing liquid hydrogen through the tube B_7, which is closed by a rubber tube with a glass stopper, the air is pumped out of the apparatus through the valve K_2. It is absolutely essential that the apparatus should be air tight, for traces of air would solidify in the liquid hydrogen and, owing to magnetic attraction, would collect in the neighbourhood of the ellipsoid.

To prevent the cooling of the upper portion of the apparatus containing the torsion spring by the boiling hydrogen, a number of large openings are made in the tube f_4 (fig. 3) arranged in such a way that no injury is done to its resistance to torsion. In addition to this copper screens surrounding f_3 and soldered to B_1, are arranged

so that the tube moves with slight torsion in them. A little cotton-wool placed on the bottom of the vacuumglass and attached to the holder lessens the sudden bubbling [1]) up of the hydrogen [1]).

Further additions of liquid hydrogen are made in the same way as the first. As a rule various series of measurements could be made with a single filling with hydrogen. The point of the vacuumglass which could not be silvered was protected by a small silvered vacuum beaker L containing liquid air. When the portion of the apparatus above the diaphragms B_{10} is again at ordinary temperature after a filling with liquid hydrogen, one can hardly notice that there is liquid hydrogen in the apparatus at all, if it is not above A_3.

In the course of time a little mist is precipitated on the vacuum tube. By surrounding the tube at A_1 with blotting paper, the moisture is prevented from trickling down between the pole-pieces. Furthermore a stream of air is directed against the tube between the pole-pieces. Hence the pole-pieces are in no way affected by the cryogenic operations.

f. The springs are phosphorbronze. This substance is non-magnetic and acquires very little permanent set. Springs of the same constant can be made by winding a spiral either of a thin short wire or of a much longer thicker one. Of the two, the one which has the greater mass will experience the smaller specific changes, and consequently will be the more perfectly elastic in working. This circumstance has been duly taken into account. The springs are provided with straight extensions in the direction of their axis and are connected with the holder and the rod k (fig. 3) by screws. The turns of the spirals do not touch each other. The temperature of the spring is measured by a mercury thermometer that is clamped against the cap D and with it is insulated with wool. The constants of the two springs used are 261000 and 22300 dyne-centimetres per radian. The corrections for the influence of the stretching wire and for the temperature change of the spring will be discussed in § 4.

The ellipsoids of iron, nickel and cobalt are 3 mm. long and 1,333 mm. thick. They have been made with great accuracy by the Société Génévoise pour la Construction d'Instruments de Physique. They were turned under a microscope giving a 30-fold magnification and provided with a camera lucida so that the image of the object and an enlarged drawing could be superposed. Measurements with

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[1]) Should this occur one must ensure that the oil of the damper is not cooled by the drops that are thrown up.

the dividing-engine have shown that the ellipsoids are very accurately shaped.

The iron was obtained by melting pure electrolytic MERCK iron contained in a magnesia boat in an electrical resistance furnace and in an atmosphere of nitrogen. The nickel and cobalt were prepared in the same way, starting with the purest possible nickel and cobalt powder specially prepared by MERCK for these experiments. The magnetite was obtained by constructing an approximate ellipsoid from a drop of very pure magnetite obtained by melting very pure MERCK sesquioxide in an oxy-hydrogen flame. Since experiment showed that it was only at very high temperatures that the last trace of oxygen was driven out and real magnetite [1]) obtained an iridium cupola was used for this operation.

Ellipsoids of approximate shape were also constructed from GOLDSCHMIDT chromium and manganese and MOISSAN vanadium. As can easily be seen it is not necessary for comparative experiments that the ellipsoids should be constructed with particular accuracy. This was, moreover, experimentally demonstrated for magnetite, of which various samples roughly worked to various ellipsoidal shapes were used for obtaining curves for the thermal change at high temperatures, and these curves were in agreement with the theoretical curve, and consequently with each other.

§ 3. *Experimental method.* As mentioned in the introduction our aim was not to obtain absolute values for magnetization in strong fields at ordinary temperature and at the temperature of liquid hydrogen, but to compare the values at these temperatures; for we might expect that the change would be only a small fraction of the quantity to be measured. Hence it was an obvious procedure to make observations at these temperatures alternately in the same field. The change, however, from the one temperature to the other necessitated operations of such duration as to prohibit the use of this method. Hence we usually began with a series of measurements at ordinary temperature, in which the field was made the required series of strengths. Then an analogous series of measurements was made at a low temperature, and after the apparatus had returned to ordinary temperature, some individual measurements were repeated so as to make sure that the apparatus had not in the meantime undergone any change.

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[1]) See also P. WEISS. Arch des Sc. phys. et nat. fevr. 1910 and Journ. de physique, 4e Sér. t. IX mars 1910.

Each series of measurements consists in turn of two branches. First by tentative approximation from both sides for all values of the field those values of the azimuth of the electromagnet are found for which the couple is a maximum. In this way two azimuths are found which are symmetrical with respect to the major axis of the ellipsoid and which exert couples of opposite sign. This determination can be made accurately to within 0,5° to 1°, which is quite sufficient. Then follows the true measurement in which the magnet without current is placed in one of these positions, the circuit is closed and immediately afterwards the deflection is read. As soon as this is done, the circuit is broken, the magnet is placed in the symmetrical position; once more the current is allowed to flow and the new deflection is obtained. Since these operations occupy only a short time, the after effects in the spring are of no account. The difference between the scale readings gives twice the value of the couple to be measured, independent of the residual magnetism remaining after the current was broken, which however occasioned only an extremely small couple. The field was given as a function of the current indicated by the ammeter. For these observations the same ammeter (SIEMENS and HALSKE instrument, no temperature coefficient) was used which was employed in the study of the field. This method of evaluating the field was quite sufficient for our purpose. The distance between the pole-pieces was read off the divided cylinders of the magnet and was verified by passing between them callipers which had been previously adjusted to the desired distance. The fields given above are corrected for the demagnetizing fields of the ellipsoids.

§ 4. *Corrections and controls; auxiliary measurements.* The inherent magnetism of the holder is not so weak that the corrections necessary for it may be neglected. On that account a series of measurements was made with no ellipsoid in the holder at ordinary and liquid hydrogen temperatures. With the weaker spring we found:

TABLE I.

Correction for the magnetism of the holder.

	ordinary temperature	$t = 20°.2$ K.
4000 gauss	0.18 cm.	0.26 cm.
8000	0.29	0.48
12000	0.36	0.61
16000	0.43	0.73
20000	0.50	0.86
24000	0.57	0.98

For the stronger spring these corrections are multiplied by $\frac{22300}{261000}$; they are very small. Direct measurements have shown that the values calculated in this way are correct, which indicates that the inherent magnetism of the carrier is not changed by the various operations of mounting.

There is still a correction to be applied to the couple-ratio for the change in elasticity of the steadying wire under the carrier when its temperature changes from ordinary to that of liquid hydrogen. To obtain that correction the ratio of the torsion modulus of the platinium iridium wire and that of the weaker of the phosphorbronze springs was measured at the two temperatures. This was done in an apparatus similar to the one we have described with the exception that the cap D could turn relatively to the cover. By a mirror method the position of the cap was read on a scale at a distance of 175.9 cm. The cap was turned through an angle of about 360°, and the exact measurement of the angle was obtained from the same scale. This angle is the sum of the torsions of the spring and the wire caused by the same couple. The torsion of the wire was read from the mirror of the holder. In this way the ratio of the modulus of the wire to that of the spring was found to be

0.0125 at ordinary temperature
0.0144 in liquid hydrogen.

The fourth decimal is uncertain; hence the correction is two thousanths for the weak spring and two ten-thousandths for the stronger. The temperature coefficient of the phosphorbronze spring was obtained from determinations of the period of oscillation of the same oscillating system while the spring was first at the ordinary temperature and then surrounded with steam. By means of the temperature coefficient thus determined viz.:

$$k = -0{,}00053$$

the observations are reduced to the same temperature.

The temperature of the liquid bath in the vacuum tube was proved to be constant to 0,1 degree, by carrying out temperature measurements with a platinum resistance thermometer placed at different heights in a similar vessel. When placed alongside the thermometer θ it indicated temperatures corresponding with those deduced from the vapour pressures.

Capillary action in the oil damper.

Care was taken to fill the oil vessel to such a height that the cylindrical ring carrying the vanes of the damper was partly immersed

in the oil so that the vanes were completely immersed and should experience no capillary action. But still we wished to know the order of magnitude of the forces brought into play by capillary disturbances; for this purpose we greatly magnified them. A damper as like ours as possible was filled only to such a height that the vanes and partitions intersected the surface of the liquid. The movable portion was suspended by a platinum-iridium wire 20 cm. long and 0,1 mm. thick; deflections were read from a mirror on a scale 2 metres away. The oil vessel was placed successively in two different azimuths such that the approach of the vanes towards the partitions would bring into play couples of opposite moments. The scale deflection was 5 cm. The moment of the couple is therefore of the order of two thousandths of that of the couple exerted on the nickel ellipsoid.

§ 5. *Details of the observations.*

Nickel.

The first series of measurements was made at 17°.2 C.

TABLE II.

H (gauss)	I^2 (cm. of the scale)
2230	89.42
6250	89.97
10270	90.12
13280	90.34
17760	90.50
20300	90.66
21540	90.79
22760	90.81

The scale reading was always corrected for the ratio of the tangent of the double angle to the double angle of the deflection. The zero as determined by the mean of readings to left and right remained constant to a few tenths of a millimeter.

After this series the apparatus was accidentally damaged; it had therefore to be taken to pieces and remounted. That occasioned a small change in the magnitude of the deflections. Since the change of I^2 with H is determined by the foregoing series, only two points were subsequently determined at ordinary temperature before and after determinations in liquid hydrogen.

TABLE III.

$t = 19°.5$ C.		Hydrogen at atm. pressure (20°.2 K.)	
H (gauss)	I^2 (cm. of the scale)	H (gauss)	I^2 (cm. of the scale)
	before	1780	93.57
16100	91.74	5410	100.49
20540	92.09	5050	101.54
	after	11830	101.84
16100	91.79	16100	102.13
20540	92.20	19050	102.34
		20540	102.51
		22020	102.48
		22840	102.49

The zero determined from the mean of readings to right and left changed by about 2 mm.

For $H = 16100$ gauss $\frac{I_{20°.2K}}{I_{19°.5C}} = 1.0549$

„ $H = 20540$ „ „ 1.0547

mean 1.0548 not connected for dilation.

Cobalt.

The measurements with cobalt did not lead to the desired result. It was the extreme difficulty of bringing the magnetization of cobalt to saturation encountered in preliminary experiments that had led to the choice of an apparatus of such small dimensions. For the other substances a weaker field would have sufficed, and hence a greater distance between the poles would have served.

In the observations at ordinary temperature something unexpected already happened. Although the mean of the readings to right and left ought to have given the zero-point of the apparatus, the point was actually observed to vary with the field. This change was afterwards seen to be about twice as great at low temperatures. The following figures bring this out clearly. (In the cobalt measurements the external field is given uncorrected for the demagnetizing field of the ellipsoid. When saturation is reached this is 5000 gauss).

TABLE IV.

Cobalt I at ordinary temperature.

H_e (gauss)	l' (cm. of the scale)	calculated zero	
4025	17.16	76.73	
8050	38.14	77.47	the observed
12075	50.48	78.96	zero was
19560	53.24	78.37	not
23340	53.29	78.18	recorded
25650	53.30	78.63	

Cobalt I at temperature of solidifying hydrogen (14°.3 *K.*).

4025	13.5	77.62	
8050	32.59	78.84	
15820	53.23	81.93	observed zero
19560	54.33	81.40	78.26
21800	54.43	81.16	
23340	54.45	81.02	
24760	54.46	80.08	

From this it appears that asymmetric disturbing forces affect the main phenomenon. It is probable that we are here dealing with phenomena of crystal magnetism arising from the fact that in the small ellipsoid the crystalline elements of the cobalt are not sufficiently numerous to realize isotropy by compensation. The magnitude and sign of these subsidiary actions are independent of the main phenomenon, and they can even be of opposite effect for both azimuths of the electromagnet; they can become of very great importance if the substance possesses a more or less pronounced magnetic plane, and the example of pyrrhotine shows us that their influence becomes greater at lower temperatures. Further, the law of approach to saturation in cobalt which differs from that which holds for the other substances is consistent with the existence of strongly developed magneto-crystalline phenomena [1]).

These experiments were repeated with a second cobalt ellipsoid, and the same asymmetric action, but somewhat weaker, was observed. But in this case a disturbance of another nature was encountered, which shows how concomitant disturbing phenomena may affect the measurement of magnetization: the magnetization at low temperature was now found to be apparently smaller than at ordinary temperature. The following table contains an extract from the results obtained with this ellipsoid.

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[1]) P. Weiss, Arch. des Sc. phys. et nat. février 1910, Journ. de phys. mars 1910.

TABLE V.

Cobalt II at $t = 18°$ *C.*

H_e (gauss)	I^2 (cm. of scale)	Calc. zero	Obs. zero
4025	20.33	77.56	
12075	54.16	76.49	77.70
23340	59.76	76.86	
25560	59.94	76.90	
Cobalt II in H_2 *at atm. pressure* (20°.2 *K.*).			
15080	53.53	76.61	
23340	58.09	77.07	78.90
25650	58.46	77.21	

The same ellipsoid was removed from the carrier and replaced with Khotinsky cement; one could easily understand that very strong strain-magnetic phenomena might be occasioned by forcibly driving it into its mount. At the same time it was for the new experiment displaced through a different angle of rotation with respect to its major axis; by this operation the sign of the change of zero point as a function of the field was reversed.

TABLE VI.

Cobalt II $t = 16°.5$ *C.*

H_e (gauss)	I^2 (cm. of scale)	Calc. zero	Obs. zero
8050	40.99	79.12	
19560	56.48	78.80	79.45
23340	57.07	78.89	
25650	57.34	78.90	
Cobalt II in H_2 *at atm. pressure* (20°.2 *K.*).			
8050	34.07	78.88	79.20
19560	53.21	78.24	
23340	54.26	78.33	
25650	54.63	78.42	

The only conclusion one seems to be able to draw from these experiments with cobalt seems to be that the increase in magnetization of cobalt between ordinary and liquid hydrogen temperatures is very much smaller than that undergone by magnetite and nickel, for, if this were not the case, the increase could not have been obscured by the disturbing influences.

ERRE WEISS and H. KAMERLINGH ONNES. "Researches on magnetization at very low temperatures."

Plate I.

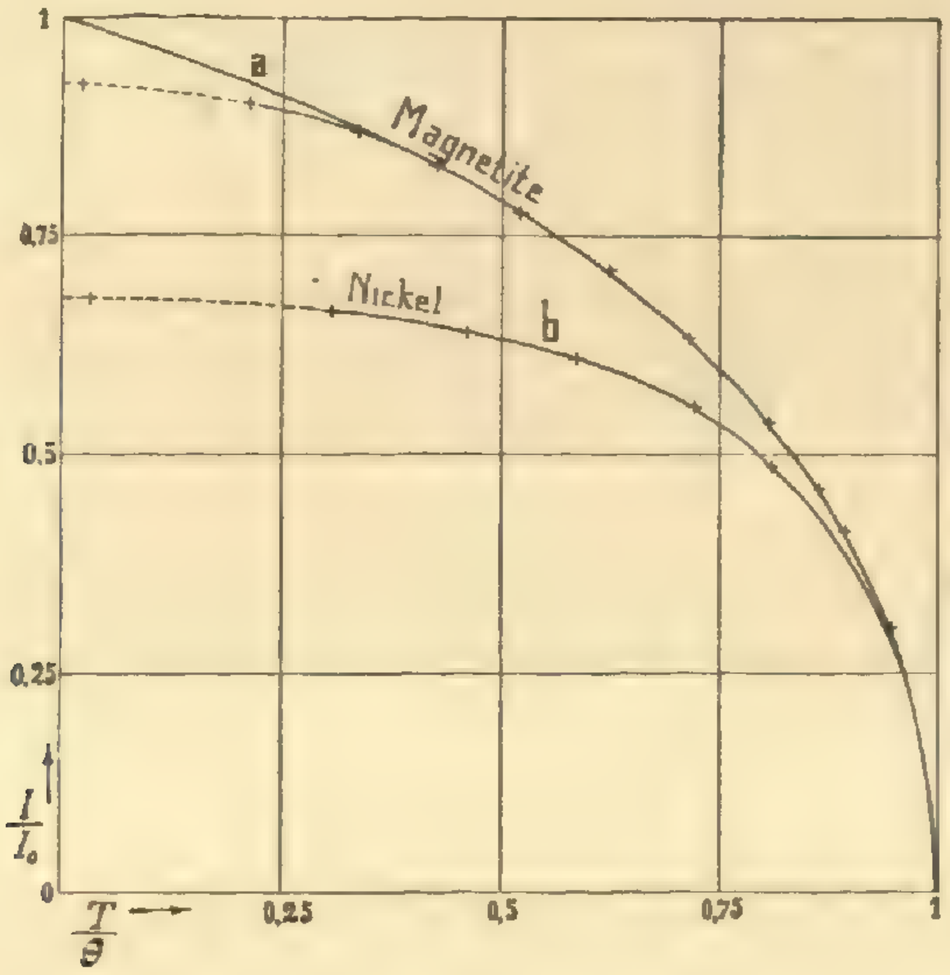

Fig. 1.

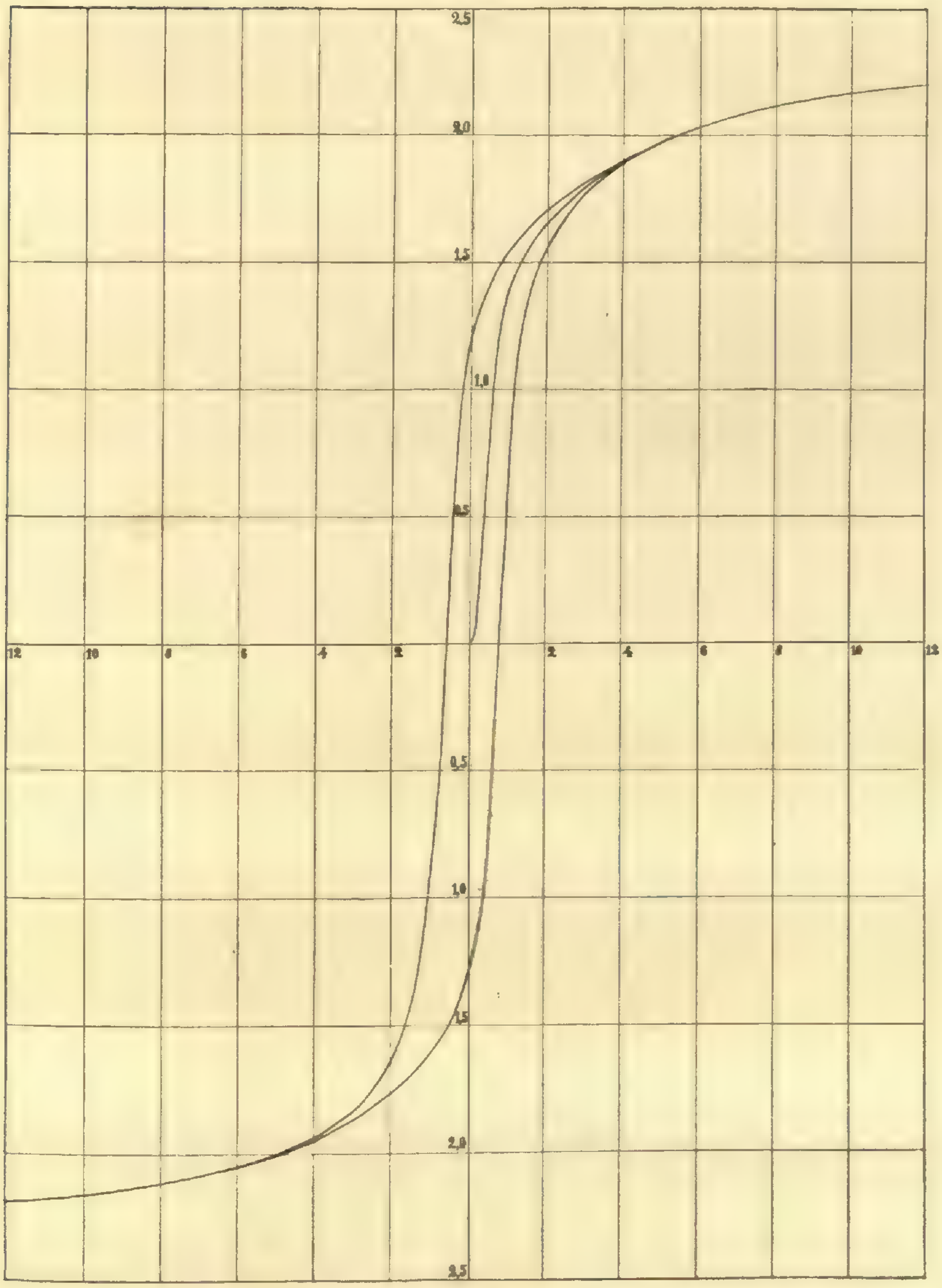

Fig. 2.

PIERRE WEISS and H. KAMERLINGH ONNES. "Researches on magnetization at very low temperatures."

Plate II.

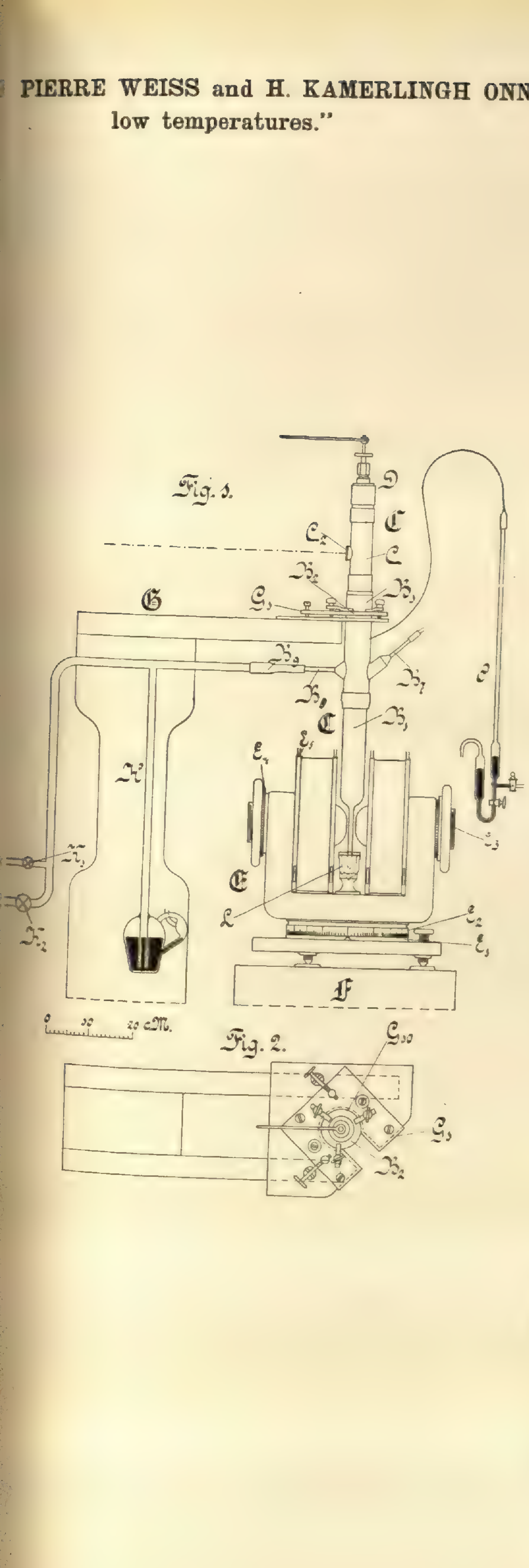

TABLE VII.

Iron.

H (gauss)	$t = 20°$ C.	I^2 (cm of scale) $T = 20°.3$ K. (H_2 atm. press.)	$T = 14°.0$ K. (H_2 solidifying)
1700	95.23	101.98	101.95
5675	98.47	102.58	
8680	98.65	103.01	
13160	98.91	103.31	
15700	99.04	103.31	103.23
16940	99.08	103.27	
18360	99.06	103.27	
19250	99.07	103.25	103.25

for $H = 19250$ $\quad \frac{I\ 20°.3\ \text{K.}}{I\ 20°\ \text{C.}} = 1.0209$

18360 1.0210

16940 1.0209

15700 1.0213

mean not corrected for dilatation 1.0210

In all the iron experiments the zero as deduced by taking the mean of readings to right and left remained remarkably constant. As a rule its displacement was only a few tenths of a millimetre in any one series, and 6 mm. in proceeding from one series to another.

The few measurements at the temperature of solidifying hydrogen are sufficient to show that nothing particular happens between 20° K. and 14° K.

Magnetite.

We have already mentioned that the preparation of magnetite by heating the sesquioxide needs an extremely high temperature if one wishes to make sure that the last traces of oxygen are removed. A first ellipsoid obtained from iron oxide that had been insufficiently heated exhibited only little more that one half of the magnetization that was expected; it showed, too, a very distinct hysteresis, which was about three times as great at liquid hydrogen temperature as at ordinary temperature, while in all the experiments with the other substances hysteresis phenomena were insignificant. Moreover, the magnetization of this substance was the same at ordinary and liquid hydrogen temperatures, while between them it reached a maximum.

These peculiarities were not displayed by a second ellipsoid cut

out of well heated magnetite, but with this second ellipsoid further phenomena were observed which have not yet been explained but which seem to be of secondary importance. The zero point deduced from the mean of the two scale readings differed noticeably from the observed zero, while in any one series of measurements at the same temperature it remained practically constant. Further — and this is more worthy of notice — the deviations differ according to the direction of the field. It would clearly be very rash to attempt to ascribe to magnetite a hemimorphous symmetry like that of tourmaline from this sole observation. It seems more probable that some experimental error has here escaped our notice, and this can the more readily be accepted seeing that magnetite gives results much less regular than those of the metals. The following table contains an extract from the observations; the observations for the positive and negative directions of the field are given separately.

TABLE VIII.

Magnetite.

$t = 15°.8$ C.

Observed zero — calculated zero + 0.9 cm.

H (gauss)	+ Field I^2 (cm. of scale)	— Field I^2 (cm. of scale)
8600	71.40	71.72
18100	71.83	72.00
21800	71.75	72.57
23300	71.99	72.57
24200	71.77	72.45
	H_2 under atm. pressure (20°.3 K.)	
8600	79.78	79.88
18100	80.69	80.79
21800	80.73	80.96
23300	80.34	81.10
24200	80.08	81.37
	H_2 solidifying (14°.0 K.)	
18100	80.90	81.10
21800	81.12	81.64
24200	80.96	81.84

From these numbers follow these ratios of the intensities at 20°.3 K. and 15°.8 C.

TABLE IX.

H gauss	Field + $\frac{I_{20°.3K.}}{I_{15°.8\,C.}}$	Field − $\frac{I_{20°.3K.}}{I_{15°.8\,C.}}$
8600	1.0559	1.0553
18100	1.0591	1.0601
21800	1.0593	1.0567
23300	1.0564	1.0572
24200	1.0563	1.0628
mean	1.0574	1.0564

Hence $\frac{I_{20°.3K.}}{I_{15°.8\,C.}} = 1.0569$ not corrected for dilatation.

Similarly for the ratio of the magnetization at 14°.0 K. to that at 15°.8 C. we find

1.0609 1.0622

hence

$$\frac{I_{14°.0K.}}{I_{15°.8\,C.}} = 1.0616 \text{ not corrected for dilatation}$$

a ratio which deviates from the foregoing in the expected direction.

Collecting the foregoing results we find in this branch of the research for the ferromagnetic substances omitting the correction for dilatation (see note 2 pg. 11)

Nickel $\frac{I_{20°.2K.}}{I_{17°.3C.}} = 1.0548$

Iron $\frac{I_{20°.3K.}}{I_{20°\,C.}} = 1.0210$

Magnetite $\frac{I_{20°.3K.}}{I_{15°.8\,C.}} = 1.0569.$

(March 24, 1910).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Saturday March 26, 1910.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van 26 Maart 1910, Dl. XVIII).

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Physiology. — "*Communications about the electrogram of the atrium cordis.*" By Dr. A. K. M. NOYONS. (Communicated by Prof H. ZWAARDEMAKER).

(Communicated in the meeting of November 27, 1909).

Involuntarily the ventricle-image, the tops R and T of which are prevailing, has, in the study of the electric phenomena of the heart, up till now been the principal subject. The top P by its smaller size drew less attention and at the outset was not even observed.

At present, however, there are in the literature already some data to be found here and there concerning the top P. Thus EINTHOVEN [1]) has pointed out how with increased action of the heart after great physical exertion P may gain in size, how under certain definite circumstances P may more or less be split up into a dim double-topped image, and besides how under pathological relations top P may be altered, which is demonstrated by cases of mitral stenosis. In this case of disease P would appear longer and enlarged, which EINTHOVEN thinks may be attributed to a stronger activity of the atrium for the sake of its compensative function. Division of the top may also appear. KRAUS and NICOLAÏ [2]) have corroborated this find, just as SAMOJLOFF [3]) and STESHINSKY [4]), who have also been able to prove that the phenomenon is not pathognomonic, but depends on the relative welfare of the heart in case of mitral stenosis.

According to VAANDRAGER [5]), top P in absolute measure would be higher with the dog than in man. With N. vagi cut through VAANDRAGER found in the dog that P grew three times its height and conversely could he make P smaller by stimulating the N. vagi. Besides this diminution he got at the same time an alteration in the form of top P.

With moderate bleeding of a sample-animal P increased in size, whilst after strong bleeding P grew smaller in the dog.

Top P was from the outset attributed by EINTHOVEN to the ventricles.

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[1]) EINTHOVEN: See: Onderzoekingen van het Physiol. Lab. te Leyden. Second Series VII and the literature pointed out there.

[2]) KRAUS F. and NICOLAÏ G. F. Ueber das Electrocardiogramm unter normalen und pathologischen Verhältnissen. Berl. klin. Wochenschr. 1907 No. 25 and 26.

[3]) SAMOJLOFF A. Electrocardiogramme. Jena 1909. Sammlung anat. und physiol. Vorträge.

[4]) SAMOJLOFF A. und STESHINSKY. Ueber die Vorhoferhebung des Elektrokardiogramms bei Mitralstenose. Münch. mediz. Wochenschr. No. 38. 1909.

[5]) VAANDRAGER B. Dissertatie Leiden 1908.

The following grounds may be adduced for this, partially borrowed from my experiments:

1. *P* always appears with a definite interval of time before the mechanical change of the atria.
2. *P* continues existing at the registration of an isolated atrium (Rana, Emys).
3. *P* is absent when the electrogram is written of the isolated heart-ventricle of Anguilla vulgaris.
4. *P* continues existing when a certain detraction of the ventricle does not show itself. This may be observed both in the pathological heartblock and in the heartblock called into existence by experimental causes, among others:
   a. by stimulating the N. vagus in the dog or the tortoise.
   b. by administering toxical materials like chloroform.
   c. by forming, resp. removing a ligature on the boundary of auricles and ventricles in Rana.
5. *P* may be made to disappear temporarily, when in appropriate sample-objects a heartblock is brought about by stimulating the N. vagus at the transition of the sinus to the atria, with which, it is true, the sinus-contractions are preserved, but the atrium-contractions with top *P* in the electrogram disappear.
6. *P* does not arise from the sinus, witness the fact that a small top may be registrated before the appearance of top *P*, which may be attributed to the sinus.
7. Size and form of *P* depend upon the way in which the atrium is derived.

If we consider the electrocardiogram of man and animal superficially, we get the impression that *P* has a very simple form. Under quite peculiar circumstances this shape has been seen to alter. On closer investigation, however, it has appeared to me that the electric phenomenon of auricles is practically a whole complex. This becomes clear at the registration of an isolated pulsating atrium. Very fit for this purpose is one of the auricles of the heart of Emys.

Thus the adjoined figure 1 renders the electrogram obtained by derivation of apex and basis of an isolated right atrium of Emys with the appertaining myogram registered by simple suspension [1]).

This image, in many respects, makes us think of an electrogram

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[1]) The registration of the electrograms was brought about by means of EINTHOVEN's string-galvanometer (EDELMANN's small model) according to the method pointed out before; see: Proceedings of the Kon. Akad. v. Wetensch. 31 Oct. 1908.

of the ventriculus cordis, as it is to be registered with numbers of animals and with man.

The tops that are found in this atrioelectrogram are, as it were, analogous to the tops Q, R and P of the ventricle phenomenon and may respectively be called here P_α, P_β, P_δ. The tops P_α and P_β, like Q and R, which are analogous to them, fall in the ventricle-image wholly before the commencement of the muscle-contraction. In the electrocardiogram of man and animals derived indirectly, we find, evidently on the ground of the P-elevation, only P_β expressed.

Again by derivation of a heart of Rana derobed from ventriculus, accordingly consisting only of sinus and atria, we get, a proportionately less large, but yet also a complicated electrogram of the atrium, as also by registration of the isolated atrium of the carp, where for example the first half of the atrioelectrogram shows a pronounced diphasic nature.

It is also possible with a sample-object to derive both the atria at the same time. The same thing I did also with the cut-out heart of Emys deprived of ventriculus, where one electrode invariably found a place on the backside of the sinus, whilst two other electrodes respectively caused the derivation of the atriatops. By means of a swing-apparatus the galvanometer was connected with the sinus and respectively with one of the atria or with both. The electrogram of the one atrium obtained in this way differs at such a derivation somewhat from the image got with the other atrium. The electrogram of the left atrium has a strong diphasic character; at derivation from the place of separation between the two atria with the back wall of the sinus, we get a less pronounced diphasic image, whilst the electrogram of the right atrium is only very feebly diphasic.

The action-current of the heart is considered as a summary utterance of the electric negativities, which show themselves in the tissue successively in different places and at different times. This negativity, as HERMANN formulated it, arises by the circumstance that every point of an irritable tissue at the moment of the stimulation stands in a negative relation to the parts that are in rest. At the derivation of such a tissue in different succeeding points we shall, therefore, every time get a deviating electrogram, but at the same time we shall be able to get an impression of the way which the proceeding stimulus has taken through the tissue. For this last purpose I have effected derivations of one atrium in different places, lying in regular order. In this experiment I made use of the heart of Emys deprived of its ventricle and then derived with an electrode constantly from the sinus, whilst the other electrode (store-electrodes with

movable pith) was placed: 1. at the apex; 2. $^1/_2$ cm. lower than the apex and 3. 1 cm. lower than the apex of the atrium. This took place both for the right and the left atrium, and also for the two atria combined.

Fig. 2 (*a*, *b*, and *c*) shows how the amplitude of top P_3 from the atrio-electrogram diminishes in size as we descend with the electrode from the apex along the lateral side of the right atrium. The greatest potential difference, therefore, is manifest between basis and point, whilst each point of the atrium, lying lower than the apex, at derivation offers a smaller potential difference with the sinus. This is quite in accordance with the usual representation, at which it is supposed that, taking into consideration the fact that the contraction-stimulus arises from the sinus, the stimulus regularly goes on in the atrium tissue from the basis to the point of the atrium.

In the same figure 2*c* may also be demonstrated the appearance of a small elevation, with following slight fall in the electrogram, manifesting itself a full second before the commencement of the myogram of the atria.

This elevation may be attributed to the sinus, among others for this reason that this elevation in size increases according as we draw nearer to the sinus.

Already in a former communication I have alleged grounds to prove the independence of the electrical phenomena of the heart with respect to the changes of form. In the atria of Emys this quality can be demonstrated very clearly.

A part of the heart of Emys consisting of only atria and sinus is, isolated, brought into a gas-chamber, and derived in one place from the sinus, in another place from one of the two atrium-tops or from both tops at the same time. In this way different combinations of derivation may be brought about. The movements of the two atria are, by means of a simple suspension, registered by the silhouette of the little levers. If 2 cm^3 of chloroform are administered which are evaporating in the gas-chamber, the mechanic movements are gradually growing smaller, so that at last they stop entirely, 11 minutes after the administration the chloroform. Also at an examination of the atria no trace of motion is to be observed, whilst however, the electric phenomena continue showing themselves periodically very clearly, through in shape they are a little more complicated than before the poisoning. In fig. 3*a* and fig. 3*b* the electrograms have been denoted as they are obtained by derivation of the sinus with the one electrode and derivation of the two atria-tops with a double other electrode. After evaporation of the chloroform by opening

the gas-chamber the atria begin to recover and after 39 minutes the electrical phenomena reach their original size again with their mechanical changes. Such poisoning-experiments can be repeated a few times without any great harm for the sample-object.

It is striking how the right atrium every time recovers first from the poisoning, and only later the left atrium begins to show mechanical changes. If the electrogram is examined during and after the poisoning, it is remarkable that the general form is not really altered here, but that the amplitude of the tops is greater than after the recovery of the poisoning, whilst the mechanical changes are altered in exactly the other way.

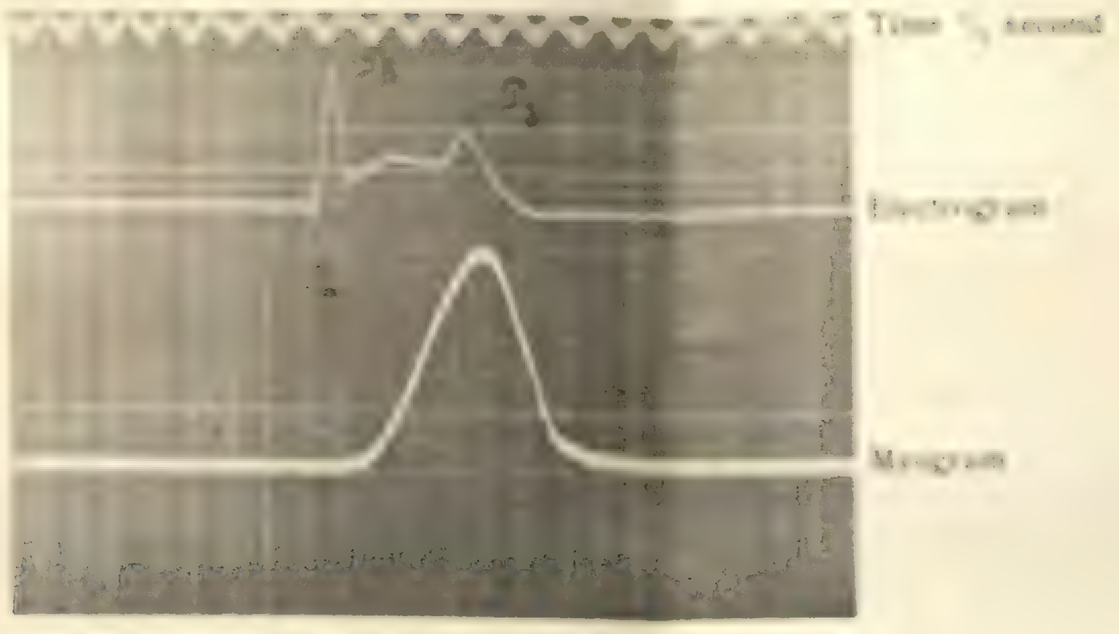
The form of the atrio-electrogram is evidently also dependent on nervous influences. EINTHOVEN and VAANDRAGER have already directed attention to this for the cardio-electrogram of a dog derived indirectly. For the atria of Emys this may be proved very distinctly at direct derivation under the influence of vagus-stimulation.

A heart of Emys derobed of ventriculus, so consisting only of the sinus and the two atria, is derived to the string-galvanometer. Derivation of one of the two atria separately or combined makes no particular difference. Therefore the derivation in the experiment takes place from sinus and right atrium-top. The right n. vagus in the neck is prepared free. The electrogram shows a fine double-topped image, accompanied by regular mechanical changes. At stimulation of the right n. vagus with induction-currents the mechanical utterance undergoes alterations. After a last contraction of the sinus, showing itself in a slight elevation in the myogram of the atria there begins for the object a vagus-standstill, which, as soon as the stimulus is put a stop to, is broken off and causes a new series of atrium-contractions with a strong sinus-contraction. During the vagus-standstill the object has produced no electrical phenomena that are to be registered. Directly after the vagus-standstill the atrio-electrograms show themselves again, but now altered in form. The double-topped image has been replaced by a phenomenon with a strongly pronounced diphasic character, which however, the stimulation being stopped, passes into the original double-topped image by a gradual alteration. In this experiment the n. vagus was stimulated by means of the sledge-inductorium of DU BOIS REYMOND, without a kernel, at a secondary coil-distance of 5.5 cm. and a LESSING-element in the primary chain.

When weaker currents are used for stimulation, a state of things may be obtained in which the vagus-standstill does not appear, but in which the peculiar alterations in the form of the electrogram of

Dr. A. K. M. NOYONS. Communication about the electrogram of the atrium cordis.

Fig. 1.

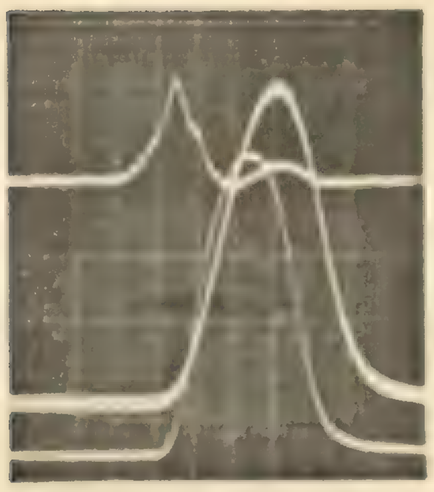


Electrogram of the isolated right atrium in Emys.

Fig. 2.

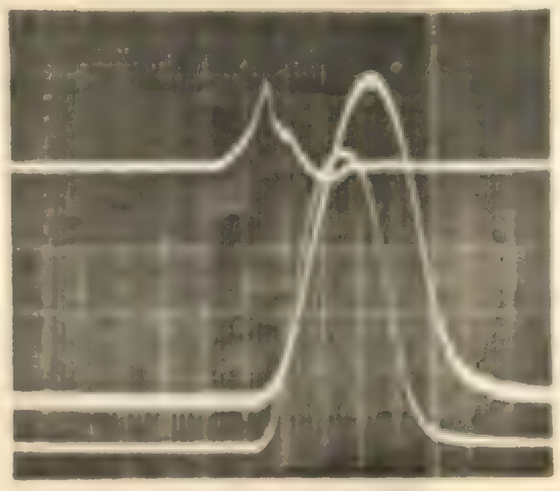
Influence of the place of derivation upon the amplitude of the atrio-electrogram in Emys.

Fig. 2a



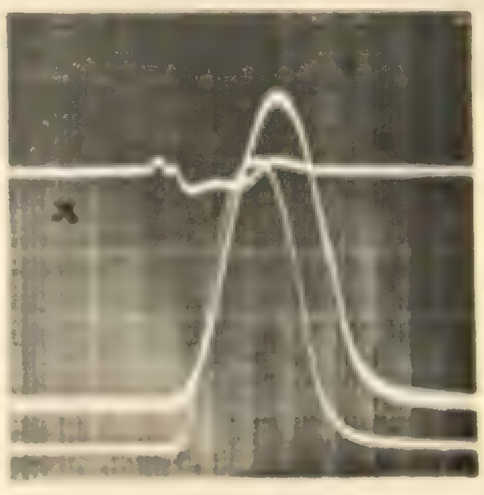
Derivation of:
sinus
and
atriumtop

Fig. 2b



Derivation of:
sinus
and
a point ½ cM. lower than the top

Fig. 2c



Derivation of:
sinus
and
a point 1 cM. lower than the top.

In fig 2 an indication of the electrogram of the sinus is found near the cross.

Fig. 3.

Independence of the electrogram with respect to the myogram in the atrium of Emys unter the influence of chloroform-poisoning.

Fig. 3a
during the chloroform-poisoning.

atrioelectrogram

atriomyogram { right / left

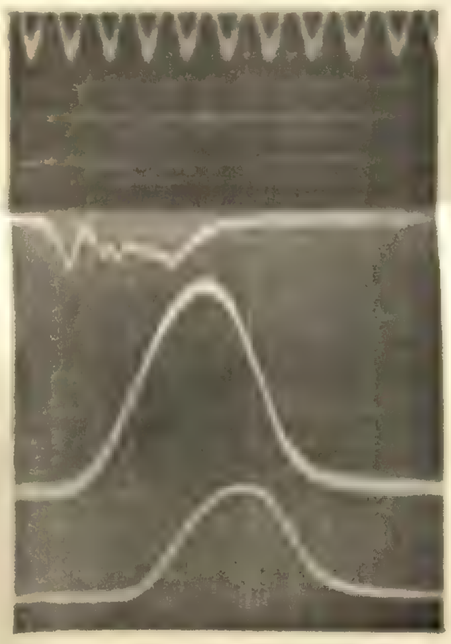
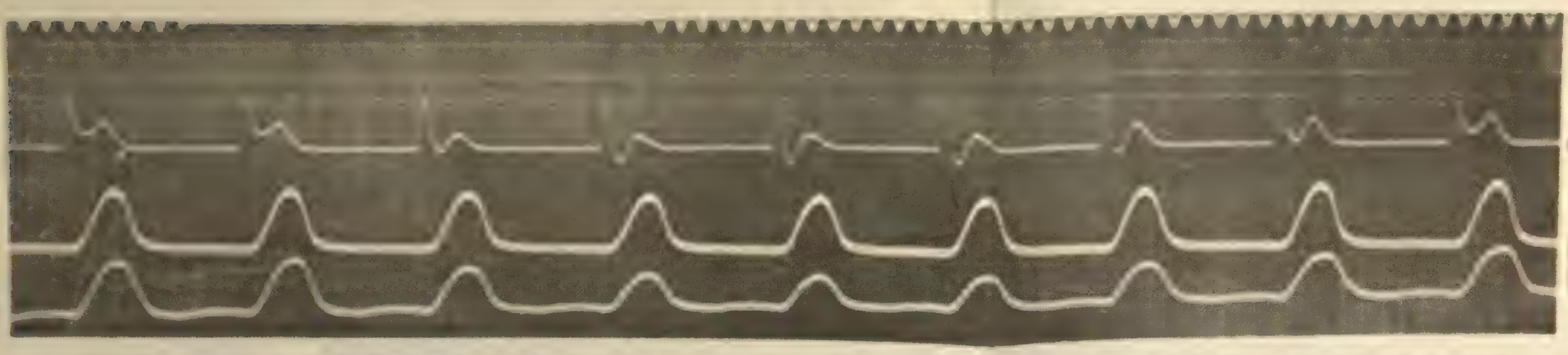


Fig. 3b
after recovery from the chloroform-poisoning.

Fig. 4.

Influence of stimulation of the right N. vagus upon the form of the electrogram of the right atrium of Emys.



Where the tonicity is interrupted the stimulation of the N. vagus takes place. The upper line of the atriomyograms belongs to the left, the lower curve belongs to the right atrium.

the atrium show themselves, as they have been described above. These alterations in the form of the electrogram, apart from tonic changes, are not accompanied by changes in the motoric utterances. Fig. 4.

The tonic alteration cannot be considered as the cause of the changes in form of the electrogram, because, in using still weaker currents as stimuli for the n. vagus, the same tonic change may appear, without having any effect on the electric utterances of the atrium.

Botany. — "*On the tests for tannin in the living plant and on the physiological significance of tannin.*" By Mr. C. VAN WISSELINGH. (Communicated by Prof. J. W. MOLL.)

(Communicated in the meeting of February 26, 1910).

In this paper a method will be described for demonstrating the presence of tannin in the living plant, a method which enables us moreover to obtain an idea of the amount of this substance in the living cells, and to ascertain whether after a given period of time the amount has increased or diminished; the method does not noticeably affect the living funtions of the plant or damage the latter to an appreciable extent.

In addition a few results of experiments on the physiological significance of tannin will be communicated; these results are in my opinion a real contribution to our knowledge of this subject.

Before proceeding to a discussion of the method which I have worked out, I think it desirable to make some observations on the meaning of the word "tannin" and to give an account of the present state of the physiological tannin-problem.

As regards the meaning attached to the word tannin there is no uniformity. Botanists formerly meant by tannin every thing in the cell which was coloured blue or green by ferric salts [1]). This has led to confusion with other substances and to the view that tannin is a generally occurring constituent of plants. REINITZER [2]) especially has drawn attention to this. As a result of his investigations he came to the conclusion that the word tannin is a misnomer, introduced into science from the leather industry. According to him it should

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[1]) J. DEKKER, De looistoffen, Bot.-chem. monographie der tanniden, 1908, V. I, p. 197 and 210.

F. CZAPEK, Biochemie der Pflanzen, II. Bd. p. 576.

[2]) F. REINITZER, Bemerkungen zur Physiologie des Gerbstoffs, Ber. d. d. bot. Gesellsch. Bd. VII, 1889, p. 187.

again disappear from scientific terminology, but his suggestion did not receive any support. WAAGE [1]) especially has objected to it. With reference to this question DEKKER [2]) rightly remarks in his botanico-chemical monograph of the tannins, that there certainly exist plant substances, which are sharply marked off from other carbon compounds by common characteristic properties, such as the property of transforming animal skins into leather, which depends on the property of forming with protein compounds insoluble in water, the adstringent taste, the presence of several phenolic hydroxyl-groups in the molecule, the power of precipitating alkaloids from aqueous solution and other properties; these substances must therefore be collected in a separate group. Until the chemical constitution of these substances is completely known, the group in which they are united, should not be split up.

Some authors, e. g. REINITZER [3]) and BRAEMER [4]) consider that a group of plant substances cannot be studied physiologically so long as our chemical knowledge of it is incomplete. WAAGE [5]), in my opinion, is quite right in not agreeing with this. Of course it will be necessary in the physiological investigation of tannins to ascertain in each case with the means at our disposal, whether the plant under investigation actually contains a substance belonging to the tannin class, so that confusion with other bodies may be excluded.

The opinion of botanists concerning the physiological significance of tannins has always been much divided. TH. HARTIG [6]) supposed that tannins contribute to the building up of the vegetable organism. SCHLEIDEN [7]) on the other hand considered that tannin is only a decomposition product of the cell wall.

In agreement with HARTIG's view tannin is, according to WIGAND [8]), a real factor in the chemical process of plant life and belongs physiologically to the group of carbohydrates, on the formation and transformation of which the life process of the plant is especially based. In contradistinction to starch, which appears as reserve ma-

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1) Th. WAAGE, Die Beziehungen des Gerbstoffs zur Pflanzenchemie, Pharm. Centralh. f. Deutschl. N°. 18, 1891, XII. Jahrg. N. F. p. 247.

2) l. c. 1908, Vol. I. p. V; Vol. II. p. 66; Vol. I. pp. 211 and 212.

3) l. c.

4) L. BRAEMER, Les tannoïdes, 1890—91. Ref. Bot. Centralbl. Jahrg. XII, 1891, Bd. 47, p. 275.

5) l. c.

6) TH. HARTIG, Entwickelungsgeschichte des Pflanzenkeims, 1858, p. 103.

7) M. J. SCHLEIDEN, Grundzüge der wissenschaftlichen Botanik, 1861, p. 141.

8) A. WIGAND, Einige Sätze über die physiologische Bedeutung des Gerbstoffes und der Pflanzenfarbe, Bot. Zeitung, 20. Jahrg. 1862, N°. 16, p. 121 and 129.

terial in the resting periods of vegetation, tannin generally belongs, according to WIGAND, to the fluid active substances necessary for growth. In some cases it appears, according to the same author, to act as reserve material. It thus follows that in WIGAND's opinion tannin is an extremely important product of vegetable metabolism. No other investigator has declared this so clearly and so emphatically.

WIGAND's view has been attacked, especially by SACHS, and has not received much support from botanists in general; this is evident, for instance, from the chapter "Die physiologische Bedeutung der Gerbsäuren" in CZAPEK's Biochemie der Pflanzen [1]), where WIGAND's view and that of TH HARTIG [2]) concerning "Gerbmehl" as carrier of tannin and organized reserve material is reckoned among the "irrigen Auffassungen über die physiologische Rolle der Gerbsäuren". WIGAND has not published the details of the observations on which his conclusions are based and this has probably contributed to the ready rejection of his results by other authors [3]).

The conception of the role of tannins arrived at by SACHS [4]) in his investigations on the germination of seeds, has received more support than that of WIGAND. SACHS considered the tannins formed in germination, to be merely excretory products, by-products or decomposition products. He thought it very improbable that tannins could serve in some way or other as material for the building up of cell-walls.

The results of some other observers agree with those of SACHS. Thus for instance KRAUS [5]), who was particularly interested in the physiological significance of tannins, arrived at the conclusion that tannin, once formed, in no case takes any further part in metabolism. According to GERBER [6]) the tannins disappear by oxidation, without the formation of carbohydrates from them. AF KLERCKER [7]) regards

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[1]) l. c. p. 588.

[2]) TH. HARTIG, Das Gerbmehl, Bot. Zeitung 23. Jahrg. N°. 7, 1865, p. 53. Weitere Mitteilungen das Gerbmehl betreffend, Bot. Zeitung 23. Jahrg. N°. 30, 1865, p. 235.

[3]) Compare EMIL KUTSCHER, Ueber die Verwendung der Gerbsäure im Stoffwechsel der Pflanze, Flora, 66. Jahrg. N°. 3, 4 and 5, 1883, p. 37.

[4]) J. SACHS, Physiologische Untersuchungen über die Keimung der Schminkbohne (Phaseolus multiflorus), Sitzungsber. d. kais. Akad. der Wiss. Wien, 37. Bd., 1859, No. 17, p. 57. Zur Keimungsgeschichte der Dattel, Bot. Zeitung, 20. Jahrg., 1862, No. 31, p. 241 and 249. Handbuch der Experimental-Physiologie der Pflanzen, 1865, p. 360.

[5]) G. KRAUS, Grundlinien einer Physiologie des Gerbstoffs, 1889, p. 38 and 44.

[6]) C. GERBER, Rôle des tannins dans les plantes et plus particulièrement dans les fruits. Compt. rend. 124, p. 1106.

[7]) J. E. F. AF KLERCKER, Studien über die Gerbstoffvacuolen. Bihang till K. Svenska Vit.-Akad. Handlingar, Bd. 13. Afd. III, No. 8, 1888. Ref. Bot. Zeitung, 47. Jahrg. 1889, p. 210.

tannins as excretion products. WAAGE [1]) calls them by-products of metabolism. BÜSGEN [2]) insists, that the observations which have been made, afford no justification for the assumption that tannin acts as a plastic material. On the other hand SCHULZ [3]) considers the tannin of evergreen leaves to play the part of reserve-material.

According to a few investigators tannins must, in some cases, be regarded as excretory products or as by-products of metabolism, whereas in other cases they take part in metabolism and serve as plastic material. Such was the conclusion of SCHELL [4]), of KUTSCHER [5]) and of WESTERMAIER [6]).

According to SCHROEDER [7]) the tannin of the birch and the maple is not a reserve material, but it is not an excretory product either. In the author's opinion it possibly in these cases constitutes a final product of metabolism. He does not, however, attempt to answer the question as to the physiological significance of tannin.

Many investigators have adopted the view, that tannins serve to protect plants against harmful external influences. These might be of very different kinds. STAHL [8]) assumes that on account of its unpleasant taste tannin serves to protect the plants from the attacks of animals, especially slugs. KRAUS [9]) also considers tannin to be a protective agent not exclusively against animals, but serving in addition to counteract the putrefaction of the plant.

In plants with evergreen leaves WARMING [10]) regards the tannin content of the epidermis as protecting the plant from desiccation, while exposed to dangerous dry winds in winter, and as being at

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[1]) l. c. p. 250.

[2]) M. BÜSGEN, Beobachtungen über das Verhalten des Gerbstoffes in den Pflanzen, Jenaische Zeitschrift für Naturwissenschaft, 24. Bd., N. F. 17. Bd., 1890, p. 59. Erläuterung zu dem Referat über Beobachtungen etc., Bot. Zeitung, 1890, p. 381.

[3]) E. SCHULZ, Ueber Reservestoffe in immergrünen Blättern unter besonderer Berücksichtigung des Gerbstoffes, Flora, 1888, p. 256.

[4]) J. SCHELL, Physiologische Rolle der Gerbsäure, Kazan, 1874 (Russian), Botan. Jahresber. III. Jahrg., 1875, p. 876

[5]) l. c p 73.

[6]) M. WESTERMAIER, Zur physiol. Bedeutung des Gerbstoffes in den Pflanzen. Sitzungsber. d. königl. preuss. Akad. der Wissensch. zu Berlin, Jahrg. 1885, 2. Halbb. p. 1124 and 1125.

[7]) J. SCHROEDER, Die Frühjahrperiode der Birke (Betula alba L.) und der Ahorn (Acer platanoïdes L.), Die landwirthsch. Versuchs-Stationen, Bd. XIV, 1871, p. 146.

[8]) ERNST STAHL, Pflanzen und Schnecken, Jenaische Zeitschrift für Naturwissenschaft, XXII. Bd., N. F. XV. Bd., p. 590 and 594.

[9]) Grundlinien zu einer Physiologie des Gerbstoffs, 1889, p. 21.

[10]) E. WARMING, Beobachtungen über Pflanzen mit überwinternden Laubblättern, Botan. Centralblatt, Jahrg. IV. Bd. 16, 1883, p. 350

the same time a means of rapidly restoring lost turgor. SCHELL [1]) considers that tannin in seeds is probably a protection against harmful influences from without. BÜSGEN [2]) also supposes that tannin affords protection to the plant.

Other authors again have attributed different functions to tannins. According to GERBER [3]) they prevent the transformation and fermentation of sugar in fruits and PFEFFER [4]) thinks it very likely, that their role also consists in fixing sugars and other substances in the cell. KUTSCHER [5]) considers it most plausible that tannin serves as a respiratory agent and is oxidized in respiration.

Various other functions have further been attributed to tannins in connexion with the metabolism of the plant. Thus WIGAND [6]) supposed that the red colouring matters are formed from tannins, a view shared by PICK [7]), MIELKE [8]), and TSCHIRCH [9]) amongst others.

Some authors connect tannins with the formation of resin. WIESNER [10]) thinks that starch and cell-wall may be transformed to tannin, and subsequently to resin. SCHELL [11]) and MIELKE [12]) also regard tannin as an intermediate stage between starch and resin and between cellulose and resin. Both authors, however, also suppose, that tannin can be converted into starch. BASTIN and TRIMBLE [13]) in their investigation of the resin-passages of conifers, have also received the impression, that tannin is connected with resin formation.

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1) l. c. p. 877.

2) l. c. p. 58.

3) l. c.

4) W. PFEFFER, Über Aufnahme von Anilinfarben in lebende Zellen, Untersuchungen aus dem botan. Institut zu Tübingen, 2. Bd., 1886—1888, p. 310.

5) l. c. p. 73.

6) l. c.

7) H. PICK, Ueber die Bedeutung des rothen Farbstoffes bei den Phanerogamen und die Beziehungen desselben zur Stärkewanderung, Botan. Centralblatt, Jahrg. IV, 1883, p. 284.

8) G. MIELKE, Ueber die Stellung der Gerbsäuren im Stoffwechsel der Pflanzen, Programm der Realschule vor dem Holstenthore in Hamburg, 1893. Ref. Botan. Centralblatt, Jahrg. XV, 1894, Bd. 59, p. 281.

9) A. TSCHIRCH, Schweiz. Wochenschr. f. Pharm. N°. 7. Pharm. Centralbl. N°. 10, 1891, p. 141.

10) J. WIESNER, Über die Entstehung des Harzes im Inneren der Pflanzenzellen, Sitzungsber. d. Wiener Akad., 1865, 52. Bd. II. Abt. p. 126 and 129. Ref. Jahresber. über die Fortschritte der Chemie etc., 1865, p. 627.

11) l. c.

12) l. c.

13) E. BASTIN and H. TRIMBLE, A contribution to the knowledge of some North Amerikan Coniferae, Amer. Journ. Pharm. 68, 1896.

According to BUIGNET [1]) tannin in fruits contributes to the formation of sugar and according to STADLER [2]) it supplies in the nectaries of *Oenothera* and *Saxifraga* the material for the formation of honey.

In connexion with the physiological significance of tannins in plant metabolism, I think it desirable to point out, what results botanists have arrived at with regard to the translocation and origin of tannins.

Some investigators suppose that tannin can be transported in the plant, namely KRAUS, MOELLER, and WESTERMAIER. According to KRAUS [3]) tannin travels as such, MOELLER [4]) thinks it probable, that carbohydrates are transported in the form of tannin compounds. WESTERMAIER [5]) leaves it an open question whether tannin travels as such, and whether starch travels in the form of a soluble carbohydrate or in that of tannin.

The opinions of botanists are divided as to the origin of tannins in the plant. As was stated above, tannin is according to SCHLEIDEN [6]) a decomposition product of the cell wall. According to TH. HARTOG [7]) it arises from starch during the germination of *Quercus pedunculata*. Similarly according to SCHELL [8]) tannin is formed from starch in the germination of the seeds of *Faba vulgaris* and *Pisum sativum*. MIELKE [9]) supposes that tannin is formed from carbohydrates, from tannin glucosides, and also from starch and from cellulose. WESTERMAIER [10]) regards it as an assimilation product but supposes that it is also formed by the decomposition of proteins. According to SCHROEDER [11]) it is formed by oxidation from organic material present in the plant. KRAUS [12]) thinks that it is a decomposition product of amido-compounds formed during the synthesis of proteins. The observations of MOELLER [13])

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[1]) H. BUIGNET, Recherches sur la matière sucrée contenue dans les fruits acides, son origine, sa nature et ses transformations, Compt. rend. 51, p. 894.

[2]) S. STADLER, Beiträge zur Kenntniss der Nectarien und der Biologie der Blüthen, Berlin 1886.

[3]) G. KRAUS, Grundlinien zu einer Physiologie des Gerbstoffs, 1889, p. 20.

[4]) HERMAN MOELLER, Anatom. Untersuchungen über das Vorkommen der Gerbsäure, Ber. d. deutschen bot. Gesellsch. Bd. VI, p. LXXX.

[5]) M. WESTERMAIER, Neue Beiträge zur Kenntniss der physiologischen Bedeutung des Gerbstoffes in den Pflanzengeweben. Sitzungsber. d. königl. preuss. Akad. d. Wiss. zu Berlin. Jahrg. 1887. 1. Halbb. p. 134.

[6]) l.c. p. 141.

[7]) l.c. p. 102.

[8]) l.c.

[9]) l.c.

[10]) Zur physiol. Bedeutung des Gerbstoffes in den Pflanzen, l.c. p. 1124.

[11]) l.c. p. 146.

[12]) Grundlinien, p. 47.

[13]) MOELLER, Mitt. des naturw. Vereins f. Neu-Vorpommern und Rügen in Greifswald, 1887.

on the leaves of *Ampelopsis hederacea* and those of BÜSGEN [1]) on germinating seeds of *Vicia Faba* and on wounded leaves which were floated on a 10 percent grape sugar solution, have proved that tannin is formed from sugar.

KRAUS [2]) and WESTERMAIER [3]) have pointed out that in some cases the formation of tannin depends on the influence of light.

It is evident from the above, that botanical opinion is much divided on the subject of the physiological significance of tannins. It may be summarized as follows. According to some botanists tannins are of no value to the plant; they are merely excretory products. Others regard tannins as protective agents against various harmful external influences. A few believe that tannins contribute to the building up of the vegetable organism. A small number think that tannins can fulfil different functions.

Various authors e.g. CZAPEK [4]) in his Biochemie der Pflanzen and DEKKER [5]) in his Botanisch-chemische Monographie der Tanniden, have pointed out that the numerous investigations on the physiology of tannins have as yet produced but few results of any importance. DEKKER arrives at the conclusion, that if this group of substances is of significance to the plant, which he thinks probable, it is quite uncertain what function they fulfil. NOLL [6]) in STRASBURGER's Lehrbuch der Botanik expresses himself in the same way. The fact that the significance of the tannins is still so obscure, is attributed to various causes. Thus according to CZAPEK [7]) a few observations of microscopical or chemical facts led to generalisations and to the construction of untenable theories. DEKKER [8]) further points to the imperfection of the methods of investigation and the one-sided use of these methods, which sometimes causes tannins to be confused with other plant substances.

In my opinion the chief cause must be sought in the want of criticism, which often impairs the drawing of conclusions. The physiological tannin problem is most certainly a very difficult problem, the answer to which will have to take into account a large number of factors. These factors are known to us to a smaller or larger extent,

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[1]) l.c. p. 34 and 35.

[2]) Grundlinien, p. 20 and 44.

[3]) Zur physiolog. Bedeutung des Gerbstoffes in den Pflanzen, l.c. p. 1117. Neue Beiträge etc. l.c. p. 128 and 133.

[4]) l. c. p. 588.

[5]) l. c. V. I, p. 220.

[6]) l. c. 8. Aufl. 1906, p. 190.

[7]) l. c. p. 588.

[8]) l. c. V. I, p. 210 and 211.

but unknown factors may also come into play. Hence it is necessary to exercise the greatest caution in drawing conclusions. In advancing an explanation of an observed phenomenon, we must consider carefully whether it is the only one possible and we must attempt to prove it in various ways by means of comparative experiments. The extent to which these precautions have been observed by no means corresponds to the complexity of the problem. As a result of a few experiments many observers have put forward certain explanations, when other explanations were equally plausible, or they have combated the opinions of other investigators, who perhaps had a more correct insight, although they were unable to adduce sufficient proof for it. Even serious investigators have made this mistake. I will illustrate this very briefly, by showing the insufficiency of the reasoning which led to the rejection of the possibility that the tannins might serve as plastic material.

As was said above SACHS does not believe that tannins can act in any way as plastic material in the formation of the tissues. This opinion he has partly supported by observation and partly by drawing what is in my opinion an erroneous conclusion. SACHS [1]) found that in the germination of seeds which do not contain tannins in the endosperm or in the embryo, tannins are formed in metabolism and primarily there, where the formation of tissue has just started. He never saw the tannins diminish or disappear during germination. In other cases, namely in that of the acorn and of the chestnut, where the embryo contains tannin, he did not observe a diminution either, but rather an increase. He made similar observations on the development of buds. SACHS concludes from the above-mentioned facts, that tannins remain in plants in the places where they have been formed, and that therefore they do not take part in the formation of tissues, for if this were the case, a diminution would have been observed. I consider this conclusion to be incorrect. Quite a different conclusion might equally well be based on SACHS' observations, namely that the frequent appearance or presence of tannins in tissue-formation shows that these substances have probably a function to perform in this process. Nevertheless I do not at all consider that SACHS has proved, that tannins remain in the places where they are formed and that they do not serve as plastic material in tissue formation. For if in the germination of seeds more tannin is formed than is decomposed, a diminution of the tannin content need not occur, and an increase

[1]) Physiolog. Untersuchungen über die Keimung der Schminkbohne, l. c. p. 111. Zur Keimungsgeschichte der Dattel, l. c. p. 246. Handbuch der Experimental-Physiologie der Pflanzen, 1865, p. 361.

may even take place. Reserve-materials like starch and fatty oils may not be assumed to participate directly in the building of the cell wall. They must first be converted into soluble substances. Now suppose that tannins also belong to this category, i. e. to such a plastic material as is present in the plant in a dissolved state, then it is not at all surprising that for the maintenance of growth plenty of this material should always be present, and that occasionally, when more of it is being produced from the reserves than is used up in the growth of the cell-walls, the tannin content increases. Anyhow it has not been proved that, because the tannin does not diminish, it remains unused at the place, where it has been formed, and that it does not serve for the building up of cell walls.

Like SACHS, KRAUS [1]) also assumes that an increase in the tannin content in germination proves, that this substance is not used up and does not serve as building material. Thus with regard to the germination of the acorn KRAUS states, as a result of quantitative tannin determinations, that not only is tannin not used up, but that its quantity even increases, so that it cannot be of service in growth.

Whereas SACHS only observed an increase of the tannin content of germinating seeds, SCHELL [2]) found in some plants an increase and in others a decrease or disappearance. In the first case SCHELL supposes, in agreement with SACHS, that the tannins are by-products of metabolism, but in the latter case he regards them as plastic material. With reference to what has already been said, it is a matter of course that I cannot either agree with SCHELL's conclusions. In my opinion it is not necessary to conclude, on the ground of an observed increase in the tannin content in some cases and a decrease in others, that tannins behave so differently in different plants. Supposing the tannin to be a plastic material in both cases, then the occurrence of an increase or decrease will depend on the quantities produced and used up. I also think it very plausible that in one and the same plant sometimes an increase, and sometimes a decrease takes place, according to circumstances.

Several botanists suppose that tannins can undergo translocation in the plant. How this might happen is still a moot point, but there can be no doubt that the possibility of translocation greatly complicates the question of the use of tannins as plastic material. The increase or decrease of the tannin content of a particular organ would then not depend wholly on production and consumption, but transport

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1) Grundlinien, p. 38.

2) l.c. p. 876.

to and from the organ would also have to be reckoned with. The mere increase or decrease of tannin in a seedling or a vegetable organ will not supply data of any value for the solution of the problem of the significance of tannin as a plastic material.

Hitherto botanists have chosen the higher plants for the study of the physiological significance of tannins. For the study of complicated vital processes and of the physiological significance of chemical constituents certain lower plants appear to me to offer advantages above those of the higher ones, the structure of which is so much more complicated. For such an investigation the thicker species of the genus *Spirogyra* seem particularly suitable. It is true that the tannin of *Spirogyra* has not yet been examined chemically, but numerous microchemical reactions allow us to conclude with a fair degree of certainty that *Spirogyra* contains in its cell sap a considerable quantity of tannin. De Vries [1]) has proved this, after abnormal plasmolysis, with various tannin reagents e. g. ferric salts, potassium bichromate, osmic acid. In addition to these reagents many others also give in the cell sap precipitates which agree completely with those caused in tannin solutions.

The advantages which the thicker species of *Spirogyra* have over the higher plants are the following. Pieces of the filaments may be examined microscopically without killing them or damaging them, and the changes in the cells can be studied in the living plant. They are particularly suited to all sorts of experiments. They are not too small to be handled and not too thick for microscopic examination. The various constituents of the cell can readily be observed under the microscope. As in the case of unicellular Algae a transport of foodmaterial from one cell to another is very probably excluded in *Spirogyra*. This important factor, which must be taken into account when dealing with the higher plants, need generally not be considered in the case of *Spirogyra*. By means of the centrifuge all sorts of abnormalities may be obtained, such as polynuclear cells, cells without nucleus, cells with a large and with a small chromatophores-mass, and even cells without chromatophores. In this way we can eliminate the assimilation process, i. e. the intake by the chromatophores of carbon from atmospheric carbon dioxide under the influence of light. With *Spirogyra* a number of comparative experiments may be made which are impossible in the case of the higher plants, and because

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[1]) Hugo de Vries, Plasmolytische Studien über die Wand der Vakuolen, Pringsh. Jahrb. f. wissensch. Botanik, Bd. 16, 1885, Heft 4, p. 575. Over looistofreactiën van Spirogyra nitida, Maandblad voor Natuurwetenschappen, 1885, Nº. 7, Reprint p. 7.

certain factors are excluded or eliminated, others may be studied with a greater chance of success.

Because the investigation of the higher plants has yielded such unsatisfactory results for the knowledge of the physiological significance of tannins I have attempted to obtain more definite results for the solution of this problem in the case of the lower plants, particularly of *Spirogyra*; to this I was led by the above considerations.

The first question to present itself was, which method would be most satisfactory. In the case of the higher plants investigators have followed various methods. Of the many reagents which give precipitates or colour reactions with tannins, ferric salts and potassium bichromate have mostly been preferred. Potassium bichromate especially, which yields with tannins a reddish brown or orange precipitate, has often been used, e. g. by SCHROEDER [1]), SCHELL [2]), KUTSCHER [3]), RULF [4]), SCHULZ [5]), MOELLER [6]) and BÜSGEN [7]). KUTSCHER made a dish with 8 sections, the colour of which agreed with that of the precipitate, but shaded in such a way that the intensity of the colour in two successive sections always differed by the same amount. This dish was used for the determination of the strength of the precipitates.

KRAUS [8]) determined the amount of tannin by means of titration with potassium permanganate or precipitated the tannin with cupric acetate and weighed the precipitated copper as copper oxide. The titration with potassium permanganate was also employed by RULF [9]).

These titrimetric and gravimetric methods cannot, of course, be applied to a small object like *Spirogyra*; moreover no method satisfied the demand which I had imposed upon myself. I desired a method which would enable me to determine the tannin content of one and the same cell at different periods, with sufficient accuracy to allow me to decide whether an increase or decrease had taken place, and this without killing the cell or harming it appreciably. The want of such a method had made itself felt in the investigation of various abnormal cells such as polynuclear and anuclear ones,

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1) l. c. p. 140.

2) l. c. p. 873.

3) l. c. p 38 and 39.

4) P. RULF, Ueber das Verhalten der Gerbsäure bei der Keimung der Pflanzen, Zeitschrift für Naturwiss. in Halle, LVII. Bd. Vierte Folge Bd. III, 1884, p. 42.

5) l. c. 227.

6) HERMANN MOELLER, Anatomische Untersuchungen über das Vorkommen der Gerbsäure, Ber. d. deutschen botan. Gesellsch., Bd. VI, 1888, p. LXVI.

7) l. c. p. 13.

8) Grundlinien, p. 61.

9) l. c. p. 42.

and cells containing many, few or no chromatophores. While I could determine the growth of such cells by measurement and could deduce from the size of the starch foci whether the starch content had increased or decreased, I was unable to obtain for one and the same cell an idea of the tannin content during the various periods of its existence. The usual reagents only permit of a single examination being made, because during it the cells are killed. I had therefore to look for another method.

I wondered whether methylene blue might perhaps satisfy the above requirements. According to PFEFFER [1]) this substance forms a compound with tannin in the living cell, and this compound separates as a fine blue precipitate. For various physiological investigations PFEFFER strongly recommends aniline dyes particularly methylene blue. Of this he says i.a. the following [2]): "In allen Fällen werden also Methylenblau und andere Farbstoffe wertvolle Reagentien sein, mit deren Hülfe, ohne Schädigung Aufschlüsse über Vorkommen und Verteilung gewisser Körper in der Zelle zu erhalten sind. Mit solcher vielseitig ausnutzbaren Methode lässt sich unter richtiger Erwägung nach vielen Richtungen hin eine Kontrole des jeweiligen Zustandes des Zellsaftes und der Veränderungen dieses im Laufe der Entwicklung erreichen." When dilute solutions are used, the penetration of methylene blue into the body of the plant and its accumulation in the cellsap continue, according to PFEFFER [3]), without any harm to life and even when complete saturation has taken place, it is still innocuous. *Spirogyra* was one of the objects with which PFEFFER experimented.

PFEFFER's experiments were repeated by me a few times with *Spirogyra maxima*, but with very unsatisfactory results. Even after several days only a slight granular precipitate was obtained in the cells, and at least the greater part of the tannin remained in solution; moreover, even very dilute solutions were found to be harmful. I cannot therefore agree with PFEFFER in praising his method of investigation, and after this disappointment a better method was sought.

Preliminary experiments were carried out on *Spirogyra maxima* with various tannin precipitants, such as alkaloids, antipyrine, ammonium vanadate and many others. Of all the substances examined, caffeine and antipyrine were found to be the least harmful, and therefore the action of these two substances was investigated more

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[1]) l. c. p. 190.
[2]) l. c. p. 191.
[3]) l. c. p. 195, 196 and 197.

closely, in order to ascertain their value for the study of the physiological tannin problem. In doing this, special attention was directed to the following points: whether the substances penetrated rapidly into the cell and the cell sap, whether the tannin was completely precipitated, and what concentration of solutions was required for this; the nature of the precipitate and whether it redissolved on removal of the precipitant, whether the strength of the precipitate corresponded to the quantity of tannin in the cells and whether the method was sufficiently innocuous. After a number of experiments with antipyrine and caffeine solutions of various concentrations, which were allowed to act for a longer or shorter time, I came to the following conclusion:

The antipyrine and caffeine solutions penetrate rapidly into the cells and in sufficient concentration produce in the cell sap a precipitate, consisting of minute grains or globules, which are in constant motion to and fro. In order to precipitate the tannin as completely as possible, it is desirable to have the antipyrine solutions not more dilute than 1 % and caffeine solutions not weaker than $^1/_{10}$ %. The greater the tannin content, the heavier the precipitate. Not infrequently the precipitate is so heavy, that the nucleus, which ordinarily can be readily discerned in *Spirogyra maxima*, cannot be distinguished at all and sometimes the precipitate is even heavier. If the *Spirogyra* filaments are placed in ditch water or in distilled water, the precipitate disappears in a short time, say in 10 minutes, and the *Spirogyra* threads are as before the experiment. No change whatsoever can be detected. If the *Spirogyra* filaments remain in the solution, the precipitate settles down and the small globules or spheres, of which it consists, gradually coalesce to larger globules, which appear perfectly colourless and may sometimes be very large closely resembling fat globules. This was generally the appearance of the precipitate after a few days. The settling down of the precipitate in the cells and the fusion of the globules to larger, purely spherical masses, proves that it is heavier than water and that it is liquid. From several data I deduce that it is not thinly liquid but viscid. The fusion to larger globules proceeds slowly and cannot, for instance, be brought about by a few minutes centrifuging. When the *Spirogyra*-cells with the globular precipitate are placed in water, the globules dissolve. Solution takes place more slowly, however, than in the case of a recently formed and still finely divided precipitate. If the preparations are placed in ferric chloride solution, instead of in water, the globules are coloured blue, while the cell sap is not coloured. It is rational to use caffeine as precipitant for this experiment, since antipyrine gives

a reddish-violet coloration with ferric chloride. Since the coloured compound is soluble and easily diffuses through the preparation, the ferric chloride-tannin reaction of the globules may also be detected when antipyrine is used, and the non-appearance of the reaction in the cell-sap may be observed, at least when the ferric chloride acts sufficiently rapidly. If the preparations are transferred from the antipyrine- or caffeine solution to a one percent solution of osmic acid, the globules are first coloured blue and soon afterwards black, whereas the cell sap remains colourless.

It is evident from the experiments with ferric chloride and with osmic acid, that the tannin is completely or almost completely precipitated by a one percent antipyrine solution and by a 0.1 percent caffeine solution, for otherwise the cell sap should have shown a blue or black coloration. If the antipyrine or caffeine precipitate, whether it be a finely divided recent precipitate or one which has fused to globules, is dissolved by placing the *Spirogyra*-filaments in water, and if ferric chloride- or osmic acid solution is then added, the cell-sap is coloured blue or black, just as is the case with cells which have not been treated with antipyrine- or caffeine solutions. When the cells finally die off in antipyrine- or caffeine solution, the globules are stained brown; their solubility in water has then decreased, but they still give with ferric chloride and osmic acid the reactions referred to.

By means of comparative experiments with antipyrine- and caffeine solutions, and various other tannin reagents, such as potasssium bichromate, osmic acid and ferric salts, with *Spirogyra* cells containing a varying amount of tannin, I was able to show that the strength of the antipyrine- and caffeine precipitates agreed with the strength of the precipitates and colorations, given by the above-mentioned reagents. For these experiments I used *Spirogyra* filaments, which had been centrifuged a few weeks before, and in which there were also all sorts of abnormal cells, such as cells without a nucleus, without chromatophores, with several nuclei etc. The tannin content of the cells of these filaments varied very much. First the filaments were treated with antipyrine- or caffeine solution and the strength of the precipitate in the various cells was noted; afterwards the filaments were placed in water, and when the precipitates had dissolved, they were placed in a solution of potassium bichromate, osmic acid or ferric chloride, and the intensity of the reaction in the various cells was noted. On comparing the various notes it was found that the strength of the antipyrine- and caffeine precipitates agreed with the intensity of reaction obtained with the other reagents, and therefore corresponded to the quantities of tannin present in the various cells.

The strength of the precipitates with antipyrine and caffeine was judged in various ways. Thus it was noted, whether the nucleus, which in normal circumstances is very clearly visible in *Spirogyra maxima*, could still be distinguished after precipitation of the tannin. Furthermore it was noted whether the suspensory threads, the chromatophores and the starch foci above and below in the cell could still be discerned. In order to judge in which cells the precipitates were strongest, the various cells were not only compared after precipitation, but it was also noted in which cells the precipitate first appeared and remained visible for the longest time after the filament had been transferred to water. I had previously found that the precipitate first appeared in the cells with the largest tannin content and that after the filaments had been placed in water, it could be observed in these cells for the longest time.

In connexion with the use which I wished to make of antipyrine- and caffeine solutions, it was very important to know to what extent these solutions are harmful to life and whether a short stay in these solutions, sufficiently long to obtain an idea of the tannin content, might be regarded as harmless or practically harmless to the *Spirogyra* filaments. I found that, if a one percent solution of antipyrine, or a $^1/_{10}$ percent solution of caffeine were used, made up with ditch water or with distilled water (a solution of such concentration therefore that all or nearly all the tannin was precipitated in the cells) and that if the *Spirogyra* filaments remained in this solution, no further divisions took place and growth was soon arrested or was stopped at once. If, on the other hand, solutions were used which were ten times as dilute, and which did not cause a precipitate in the cells, it was found by comparative experiments with *Spirogyra* filaments in ditch water or in distilled water, that growth was retarded by antipyrine and by caffeine, and that fewer nuclear and cell divisions occurred.

I made some experiments with a one percent antipyrine solution and with a $^1/_{10}$ percent and a one percent solution of caffeine, in order to see whether a daily sojourn of 10 minutes in these solutions was harmful to *Spirogyra*, grown in ditch water. A period of 10 minutes was selected because it is sufficient for an examination of the tannin content. The result of these experiments was, that it could not be ascertained with certainty whether the procedure employed was harmful to the *Spirogyra*. Sometimes the growth of the controls in ditch water was the stronger, sometimes that of the filaments which had been periodically treated with antipyrine and caffeine solutions. It is not improbable that the differences observed depended

largely on the nature of the cells under investigation. I surmise this because the growth of cells in normal and equal nutrient solutions also showed differences. We may deduce from the results that in general a short daily stay in the various solutions has at most a slight influence on the growth and the vital processes of *Spirogyra*. The above method of investigation of the tannin content may therefore be strongly recommended, especially when it is desired to examine the same cells repeatedly at intervals, without harming them.

As far as I have been able to ascertain, antipyrine- and caffeine solutions have not yet been employed as microchemical tannin reagents. For the sake of completeness I point out, however, that such solutions have already been used by botanists in microchemical investigation, namely by LOEW and BOKORNY [1]), to demonstrate the presence of non-organized active protein in the living cell. The above-mentioned reagents are supposed to separate this in the shape of small globules, called by these authors proteosomes. This is therefore an explanation of the phenomenon produced by antipyrine or caffeine in the living cell, which is totally different from that given by myself. As a result of my investigations described above, I adhere to my opinion that antipyrine- and caffeine solutions are valuable tannin reagents, and suppose that LOEW and BOKORNY have given an inaccurate explanation of the phenomenon which they observed.

In the historical survey I pointed out, that, as regards the physiological significance of the tannins, there is a great difference of opinion among investigators, and that in the opinion of various botanists, there is but little, which may be regarded as sufficiently proved, so that we are here face to face with a problem, which has in no way been solved. As was stated above the view that tannins might serve in the formation of cell walls has received little support and met with much opposition. With the aid of the method I have worked out, I have now been able to bring to light facts concerning *Spirogyra*, which indicate that tannin plays an important part in the formation of cell walls, and that during this process tannin is used up, so that it very probably serves as building material. Below I will mention some observations which relate to this. They refer in the first place to the conjugation.

Cells which showed a tendency to conjugate, I found to be richly provided with tannin. I could make out, that the tannin content diminished during conjugation and in the adult zygospores which were filled with reserve material, I could only occasionally observe

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[1]) O. LOEW and TH. BOKORNY, Versuche über aktives Eiweiss für Vorlesung und Praktikum, Biologisches Centralblatt, 1891, XI, p. 5.

a feeble tannin reaction with ferric chloride. It does not result from this observation what is the fate of the tannin, but when the conjugation is followed in greater detail, it is found that there is good ground for supposing, that at least a portion of the tannin serves as plastic material for the cell wall. Conjugation is a process which proceeds in such a way as to allow us to expect that its study in connexion with the point of investigation referred to will furnish us with important data, for conjugation does not start simultaneously in all cells. Some cells are in advance of others; in a smaller or larger number of cells there is evidently a tendency to conjugate, but the conjugation does not succeed, and other cells again do not show a trace of the conjugation process. Whereas the conjugating cells form much reserve material as starch and fat, those which do not conjugate are apparently very poor in contents and they finally perish. The above mentioned differences seem to be determined by accidental circumstances such as the coming into touch with cells of other filaments, the proximity of such cells and the position of the cells with regard to each other. They may even be observed with material which before conjugation consists exclusively of healthy normal cells.

The point of interest in connexion with the tannin problem is the possibility of comparing, in conjugating *Spirogyra* filaments, cells which a short time before were quite equal and afterwards show more or less important differences, induced by accidental and rather superficial circumstances. It is of interest to trace in these various cells what happens to the tannin content. This was investigated with the caffeine- and antipyrine solutions I have recommended, and it was striking to note, how differences in the development of the cell wall corresponded to the quantity of tannin present in the cells. Thus I could ascertain, that in cells where the lateral protrusion and mutual fusion had taken place, the tannin content was always appreciably smaller than in cells which only showed the first beginnings of the lateral protrusion. These two kinds of cells only differed as regards cell wall and tannin content; for the rest they still agreed perfectly. They were distributed promiscuously over the filaments, as is usual in conjugation. These facts seem to me to prove that there is a connexion between formation of the cell wall and the tannin content, and the supposition, that tannin serves as plastic material for the cell wall is very plausible.

Furthermore there is a remarkable increase in the tannin content of those cells which have not had an opportunity of conjugating or in which the process was interrupted at an early stage; these cells

degenerate and are generally described as having a poor cell content. These cells continue to produce tannin for some time and since the tannin in them is not used up in the formation of cell walls or reserve material, the tannin content increases and on the death of these cells a considerable quantity of plastic material in the form of tannin is lost.

The loss of tannin in nature, e. g. in the fall of leaves in autumn, has repeatedly been used as an argument for the view that tannin cannot be a plastic material and does not take part in metabolism. I cannot share this view and do not think the waste of quantities of a substance, which certain plants require for their development, to be at all strange, and certainly not a proof that it cannot serve as plastic material in the development of the plant. How often do things in nature fail to attain their end and how many are not wasted without being able to fulfil their purpose! Moreover, it seems to me desirable that the plant should have an excess of plastic material at its disposal, in order that development may never at any time be hindered for want of it. The fact that in the autumn the stem is unable to take up all the tannin from the leaves, or all that remains in the leaves from former abundance, hardly proves that tannin cannot serve to build up the tissues. Still less need we wonder at the waste of tannin in *Spirogyra*, for evidently it is here not the intention of nature that it should be wasted. Nature ensures a sufficient supply of tannin in *Spirogyra*, because this substance is required in development, as for instance in conjugation and spore-formation. The occasional failure to conjugate, as a result of which then much tannin is lost, does not prove that it is a waste product and not a plastic material.

A second series of observations, which show that tannin plays a part in the formation of the cell wall, relate to the formation of transverse walls. On investigating *Spirogyra* filaments containing cells undergoing division, it at once struck me that the tannin content of these cells is somewhat smaller than that of other cells, not undergoing division. The difference was not large and perhaps, even escapes detection by some of the tannin reagents which have been used hitherto, such as ferric salts and potassium bichromate, but with antipyrine- and caffeine solutions the existence of a difference in the tannin content could be established with certainty. Not only was it clear that the precipitate with antipyrine- or with caffeine solution was somewhat less in the cells undergoing division than in the others, but on treatment of the filaments with these solutions, it was also found, that the precipitate appeared somewhat later in the cells

in process of division and that on transferring them to distilled water or to ditch water the precipitate also disappeared somewhat sooner. For the sake of completeness I further mention, that no difference could be traced between the tannin content of cells in which the nuclear and cell division had just started, and the tannin content of cells not undergoing division, but the tannin content was found to have diminished, when the process of nuclear and cellular division was at its height or could be considered at an end.

These results show, that a connexion must be looked for between the diminution of tannin content and the process of nuclear and cellular division. This process really consists of two processes, going on simultaneously, and therefore the question arose, which of the two exerted its influence on the tannin content. With reference to this question I carried out some experiments.

As has already been stated, the growth of the cells and the division of cell and nucleus is stopped in a one percent antipyrine solution or in a 0.1 percent caffeine solution. I therefore studied the effect of these solutions on the formation of transverse walls and on karyokinesis, when the dividing cells and those showing the very earliest signs of the process of nuclear and of cell division, were placed in these solutions for some time. Filaments, in which such cells occurred, were left for $1\frac{1}{2}$ hours in the above mentioned solutions, and were then examined next day with regard to the division of cell and nucleus. The transverse walls, in process of formation, had been disturbed in their development, and therefore in these cases the cell was incompletely divided. The result in the cells which were on the point of dividing, when placed in the antipyrine- or caffeine solution, was more interesting; often in these cells no trace of a transverse wall could be found next day. The process of cell division had been completely suppressed.

The process of nuclear division was however quite different. In all the cells where it was going on, or where it was about to begin, it had continued to the end and two normal daughter nuclei always resulted, which were generally situated a little apart in the axis of the cell.

It follows from these experiments, that a temporary fixation of the tannin by antipyrine or caffeine prevents the formation of transverse walls, but does not directly affect nuclear division. On the strength of this result I feel justified in assuming that there must be a connexion between the diminution of the tannin content, referred to above, and the formation of transverse walls. Both abolition of transverse wall formation through fixation of tannin and the dimi-

nution of the tannin content during the formation of transverse walls, point to the tannin being necessary for, and used up in the formation of transverse walls.

In order to obtain still greater certainty with regard to this conclusion, the influence of antipyrine and caffeine on the formation of transverse walls in *Cladophora* was investigated. With ferric chloride, osmic acid, and antipyrine I did not obtain tannin reactions in *Cladophora* and I therefore was interested in knowing how, for instance, the formation of transverse walls would be affected by transferring to a one percent antipyrine solution. I found that transverse walls, which were just beginning to be formed, continued to grow until they were completed. This was even the case if the specimens were left in antipyrine solution during the whole of the process of cell division. This result still further strengthens my view that in *Spirogyra* the interruption or prevention of transverse wall formation is wholly due to the fixation of the tannin. For in *Cladophora*, where no tannin can be used in the formation of transverse walls, a one percent solution of antipyrine does not bring about this disturbance. The only ready explanation which, in my opinion, can be given of the results obtained in the conjugation and transverse wall formation, is this, that the tannin serves as plastic material in the building up of the cell wall.

I wish to add a few results to those already mentioned, which point to a connexion between the tannin content and growth of cell wall. In *Spirogyra* filaments cells are sometimes observed, which, judging from the position of the transverse walls, are distinguished from the others by increased turgor. These cells are generally also distinguished by a larger starch content. On closer examination it is found that the growth of these cells is less than that of the others, or that growth has completely come to a standstill. These symptoms indicate a pathological condition, for generally I was able to ascertain that the above-mentioned cells did not divide further and died off. I cannot give the reason for this condition, but it is remarkable that the tannin content of these cells as revealed by antipyrine or caffeine solution, is larger, and often much larger, than that of the other cells. Once more it is found, as in the case of cells in which conjugation failed, that a cessation of growth is accompanied by an increase in the tannin content.

As was shown by the investigations of GERASSIMOFF [1]) and of

[1]) J. J. GERASSIMOW. Ueber den Einfluss des Kerns auf das Wachstum der Zelle, Separat-Abdruck aus Bull. d. l. Soc. Imp. des Nat. de Moskou, 1901, No. 1 en 2, p. 193. Zur Physiologie der Zelle, Separat-Abdruck aus Bull. d. l. Soc. Imp. des Nat. de Moscou, 1904, No. 1, p. 7.

myself [1]), the growth of cells without nuclei is very slight and gradually stops completely. In anuclear cells with chromatophores and in those without chromatophores, the two kinds being obtained by centrifuging the cells before or during karyokinesis, the tannin content after a time becomes very considerable, as shown by examination with caffeine- and antipyrine solutions. In the absence of a nucleus growth stops, and as a result the consumption of tannin must have fallen off or has stopped altogether. Its production is however continued for some time; hence the increase of the tannin content in cells without nucleus. In this case also there is cessation of growth and an increase in the tannin content.

The results obtained with non-growing nucleated and with non-nucleated cells, agree with those which I obtained with cells conjugating and undergoing division, but are of less importance for the explanation of the physiological significance of tannin, because non-growing nucleated cells must be considered diseased, and those without nuclei are very abnormal. The results obtained with conjugating cells and with cells undergoing division, I consider on the other hand of great importance for the explanation of the physiological meaning of tannin, which in my opinion must be regarded in *Spirogyra* as a substance which serves in the formation of the cell walls. The tannin is here not a reserve-material, however; it belongs to the soluble substances which the plant continually requires for its development. It disappears and gives way to reserve-materials, when the plant forms zygospores and passes into the resting condition. Hence I have arrived at a result, which agrees with the conclusions published by WIGAND nearly half a century ago, but which militates against the view of later investigators, such as SACHS, KRAUS and others. For the sake of clearness I must add, that I do not at all claim that tannin is the only substance, which is used in the formation of the cell wall of *Spirogyra*, nor do I wish to argue that the only physiological significance of tannin is its use as a plastic material.

This paper is a preliminary one. It is my intention to report at some future time more fully on the physiological significance of tannin in *Spirogyra*, and to illustrate with tables the conclusions relating to the comparative experiments on the growth of *Spirogyra* filaments under various conditions, i. e. in antipyrine- and caffeine solutions, in ditch water etc. At the same time various points of investigation, relating to the tannin problem, and not mentioned in this paper, will be dealt with.

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[1]) C. VAN WISSELINGH, Over wandvorming bij kernlooze cellen. Reprint from Bot. Jaarb. Dodonaea, Vol. 13, 1904, p. 5 and 6. Zur Physiologie der Spirogyrazelle, Beihefte zum Botan. Centralblatt, Bd. XXIV, Abt. I, p. 170.

Physiology. — *"The Camera silenta*[1]*) of the Physiological Laboratory at Utrecht"*. By Prof. H. ZWAARDEMAKER.

(Communicated in the meeting of February 26, 1910.)

The extension of the means of communication calls forth nearly everywhere to a higher or lower degree the disadvantages connected with the continual presence of noise. Therefore we want in many instances apartments free from sound, and that at first in those cases in which the continuous existence of disturbing sounds forms an insuperable impediment. Such cases present themselves:

a. in acoustic experiments when the observations have to take place in the proximity of the minimum perceptibile;

b. in public consulting rooms for diseases in the ear where through the coming and going of patients the required silence never reigns, and more frequent visits render every minute investigation well nigh impossible, consequently cause also uncertainty of diagnosis, of advice and of decision in case of examination;

c. in modernly built hospitals, which with their smooth walls, naked floors, construction of stone and iron, etc. show a kind of strong resonance, and which, through their many technical 'institutions' can never be quiet; the consequence is the impracticableness of a really efficient percussive and auscultatory examination.

Since 1904 a camera silenta ($2.28 \times 2.28 \times 2.20$ M.) has been used for the purpose mentioned under *a* in the Physiological Laboratory at Utrecht[2]) and also since that time my advice has repeatedly been asked in the building of new laboratories, polyclinics and hospitals in this country and elsewhere. In connection with this I venture here to pronounce the conviction that an apartment free from sound, intended for one of the three above mentioned purposes, will have to satisfy three conditions in order to preclude disappointment. These conditions are:

1. The inner surface of the apartment has to be covered with

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[1]) Silentus, adj. occurring in GELLIUS, in a fragment from LAEVIUS used by "loca", is, on account of its shortness, preferable to silentiosus.

[2]) Ned. Tijdschr. v. Geneesk. 1905, Part I, p. 571. Zeitschr. f. Ohrenheilk. Bd. 54, p. 247.

a material that does not reverberate sound; for if this is neglected, not only the involuntary sounds that are made by us, will have a disturbing influence, but we shall also be hindered by the small remainder of sound that might still be left on account of incompleteness in the construction; the resonance of the space that is shut off will itself seize definite parts of the small quantity of noise that arises or penetrates into it and make them audible in a higher degree.

2. The isolation must be brought about by a double wall, with interstices of air of such a trifling thickness that resonance of audible tones is quite out of the question and moreover no other contact is left between the two walls than of a few narrow lead-contacts.

3. The isolation of the outer wall of the building and of its bottom has to be as complete as possible: the first isolation has to take place through a purposely constructed secondary apartment.

The first condition is fulfilled in our laboratory by means of a covering of horsehair some centimeters thick (trichopièse), as it is used in telephone-cells. Thanks are due to Dr. Biltris of Gent for making me acquainted with this material, which, moreover, procures an excellent isolation of sound.

The second condition is satisfied at Utrecht by making use, in fastening the trichopièse, of a wall of porous stone and by constructing outside it a second wall, consisting of corkstone of German manufacture. Plates of peatmoss from Klazienaveen in the province of Drente would have answered the purpose even better.

The third condition requires the exclusive use of lead-contacts. Especially the bottom has to be well provided for. At Utrecht faults have been made in this respect, which could only partly be made up for by the subsequent addition of an extra-covering.

Taking the above-named chief conditions for granted, we shall have to answer the question, whether an apartment free from sound will have to be constructed underground, on a level with the ground or on a higher floor. My answer is decidedly on a higher floor, for the conduction of the sound coming from the bottom is the obstacle which it is most difficult to overcome. An efficient isolation of the bottom can much more easily be brought about on a higher floor than on a foundation. In the first case the only thing one has to do is to provide lead-contacts with the stone beams, which in their turn are not directly connected with the bottom, whilst in the second case, under the most favourable circumstances, short columns con-

sisting of many strata can be made use of, which, however, have a constant direct communication with the ground.

As to the different tones, the most difficult thing appears to be to keep away the low tones. Inaudible vibrations of very slow periodicity are even not at all excluded in our camera silenta, so that a sensitive microphone, conducted to a gold-thread string-galvanometer does not appear to subside, not even when at a complete adaptation of the organ of hearing not a trace of sound is to be observed. (This does not disturb acoustically, but a somewhat faster periodicity would have been a hindrance).

Besides an apartment free from sound ought to have porous walls, for if perfectly impermeable walls are chosen, it will appear that in case of long experiments a ventilation is necessary, which in its turn would require the supply of ventilation-channels, consequently of sound-leaks. For double-door and double-window (the latter in my opinion hygienically indispensable) as a matter of course apparatus are wanted which require much care and a lasting control. When acoustic experiments are made, the supply of sound should come from sound-sources placed outside the apartment, right through a leaden stopper, that the principle that the two walls of the double wall should have none but a lead-contact, is not discounted [1]). Electric light, telephone, supply of air for organ-pipes and sirens through a narrow leaden tube and the necessary conducting-wire to the galvanometer offer no technical difficulties.

An accidental additional advantage of an acoustic apartment with a double wall, double door and double window, duly separated from the outer-walls of the building by means of by-apartments, is this, that it forms a calorimeter. The camera silenta at Utrecht remains without an inhabitant of a constant temperature to within 2 decigrades. By covering the trichopièse-walls with some meters of extremely fine brasswire (0,1 mm.), a bolometer may be made with a Wheatstone bridge and galvanometer placed in a by-apartment, by which bolometer the rise of temperature that the space undergoes through an inhabitant, may be measured. The production of heat which this causes is determined empirically (D'ARSONVAL). As a respiration-calorimeter, however, the sound-free apartment is not to be used. This is impossible because the walls are porous, and if this is given up, it is no longer free from sound for longer experiments.

A number of investigations may take place in the camera silenta.

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[1]) The leaden stoppers are 5 cm. thick and possess a central bore, at its narrowest point being 0.4 cm. wide; comp. Onderz. Physiol. Lab. Utrecht (5) VI. p. 138.

Those which have been made in the last six years, are, it is true, not so numerous and extensive as I should wish, but an enumeration with a list of the publications may follow here in order to serve as an example of what is to be reached in a sound-free apartment.

1. The sensation of stillness may be experimented on; unless a perforation of the tympanum exists, a kind of buzzing may be observed, in which at a closer analysis a soft rustling as of the wind in the tops of the trees, accompanied by a high-toned whistling ($\pm g^6$) may be distinguished; persons in whom this physiological ear-buzzing is indistinct, perceive a feeling of oppression [1]).

2. The influence of the adaptation may be traced; then appears among others a gradual diminution of the physiological tinnitus aurium, which after a 3 hours' stay in the sound-free apartment has entirely disappeared (BORTOLOTTI), whilst at the same time the feeling of oppression, if existing, gradually increases (MINKEMA); from this one might be inclined to derive that the physiological ear-buzzing, entirely or partly, possesses the character of an after-image [2]).

3. The phenomenon of accommodation, discovered by HENSEN, may be more closely studied, by conveying to a person standing outside the camera silenta through bone-conduction the tone of a tuning-fork, which then from the person's ear is conducted into the apartment through an auditory tube; whenever a metronome placed outside the apartment is ticking, the sample-person accommodates and the observer bears a strengthened sound (QUIX).

4. From the shortest exposition-time the smallest observable number of sound-vibrations may be derived in the tone of a tuning-fork or that of an organ, conducted to it from the outside; according to BODE this number seems to vary in the scale in a typical manner (DE GROOT [3]) and VAN MENS).

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[1]) For my ear the physiological ear-buzzing can be suppressed: *a.* by the ticking of a watch; *b.* by the sound of a tuning-fork of the c'-pitch and a sound-force of 68.10^{-3} Erg. per cm.^{2} and per sec. (Erg. d. Physiol. 1905 p. 452).

[2]) According to BORTOLOTTI the buzzing returns directly, after one has left the camera for a moment and then returns.

[3]) H. DE GROOT, Ztschr. f. Sinnesphysiol. Bd. 44 p. 18 and Onderz. Physiol. Lab. (5) X p. 137.

5. The minimum perceptibile during the unity of time may be fixed by the scale (MINKEMA [1])).

6. The limit of distinction may be traced and the typical variation it undergoes in the scale (DEENIK [2])).

7. The sensation of a report, observed by HENSEN at a sudden intonation or interruption of siren-tones, may be demonstrated in tones of different origin and pitch, with the aid of a sudden opening or closing of a telephone-contact or a sudden opening or closing of a particularly constructed lead cock.

8. The spreading of the sound round a tuning-fork with the situation of the well-known interference-planes may be accurately traced, without making the mistakes that must necessarily arise in apartments with echoing walls.

9. The action of the winding mollusc-shells as to their resonance for buzzes may be proved directly.

10. The sound-extinguishing action of different means of isolation may be traced with perfect security; for reports by dropping steel balls on a steel plate [3]) (fall-phonometer of ZOTH), for tones by electrically touching purely tuned bells; in both cases the instrument put in a small non-resonant space; the walls of this space are covered with the materials that are to be examined, and, on the one side the energy with which the bells are touched, and on the other the distance at which the sound is heard, is defined; the completest isolation with an equal thickness of the walls is got in the case of trichopièse, then follows the peatmoss-plate from Klazienaveen, then the corkstone; other materials that we examined had a considerably smaller sound-extinction.

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[1]) H. F. MINKEMA, Onderz. Physiol. Lab. (5) VI. p. 134.

[2]) Meeting of this Academy 3 Nov. 1905.

[3]) In order to prevent resonance the stale plate has to be soldered upon a heavy piece of lead.

Mathematics. — *"On pairs of points which are associated with respect to a plane cubic."* By Prof. JAN DE VRIES.

(Communicated in the meeting of February 26, 1910).

1. By the symbolical equation

$$a_x^3 = 0$$

a plane cubic c^3 is represented. If the points X, Y, and Z are connected by the relation

$$a_x\, a_y\, a_z = 0,$$

each of them lies on the (mixed) polar line of the other two, and every two of those points are harmonically separated by the polar-conic of the third point; they form a polar triangle of c^3.

Let us look more closely at the case that the three points lie in one line l; then Z is the point of intersection of l with the polar line of X and Y.

It is evident that the triplets X, Y, Z lying on l form a cubic involution I_2^3 of order two having the points of intersection P,Q,R of c^3 with l as threefold elements.

According to a well known property of the I_2^3 we find that P, Q and R form at the same time a group of the I_2^3. This is indeed directly to be seen; for, the polar conic of P intersects l in P and in the point H, which is harmonically separated by Q and R from P; the polar line of Q with respect to that conic therefore passes through R.

To I_2^3 belongs a neutral pair, U, V forming with each point of l a triplet and therefore having l as polar line. The polar conics of the points lying on l form a pencil; two of those conics u^2 and v^2 touch l in the points V and U.

We shall call U and V *associated points*.

Evidently each point U is associated with two points V, viz. with the points which have the polar line and the polar conic of U in common. The associated pairs are thus arranged in an involutory correspondence (2, 2).

If l becomes tangent to c^3, then in the point of contact L two threefold elements of the I_2^3 unite themselves with the two neutral points U, V. For, all polar conics whose poles lie on l pass through L and one of those curves touches l in L. So c^3 is curve of coincidence of the (2, 2) correspondence.

2. We shall see whether there are points U, for which the corresponding points V form again an associated pair, so that there is a triplet of points which are two by two associated. If we take the three points as vertices of a triangle of reference, their polar conics will be represented by:

$$a_1 x_1^2 + b_1 x_2 x_3 = 0, \qquad a_2 x_2^2 + b_2 x_1 x_3 = 0, \qquad a_3 x_3^2 + b_3 x_1 x_2 = 0$$

for, each of those points has the connecting line of the other two as tangential chord with respect to its polar conic.

If $u = 0$ is the equation of c^3, the three polar conics are also represented by

$$\frac{du}{dx_1} = 0, \qquad \frac{du}{dx_2} = 0, \qquad \frac{du}{dx_3} = 0.$$

From this ensues in the first place that the coefficients b_1, b_2, b_3 must be equal. Farther on it is directly evident that the equation of c^3 is:

$$a_1 x_1^3 + a_2 x_2^3 + a_3 x_3^3 + 3\, b\, x_1 x_2 x_3 = 0.$$

The triangle of coordinates is therefore a triangle of inflection, i. e. a triangle of which each side contains three points of inflection of c^3. There being four triangles of inflection, the (2,2)-correspondence of the associated points contains *four involutory triplets*.

3. We shall now determine the locus of the associated pairs, collinear with a given point D.

In the first place D is a node of the locus; the points D' and D'' associated with D are the points of intersection of the polar conic d^2 of D with the polar line d of D. The locus is therefore a nodal biquadratic curve d^4.

The tangents out of D to c^3 are at the same time tangents to d^4, for in their tangential points two associated points continually coincide.

So d^3 is the conic of BERTINI of d^4. For an arbitrary nodal c^4 this conic contains besides the six points of contact of the tangents out of the node, the points of intersection of c^4 with the line connecting the two tangential points of the node, and the tangents in those "fundamental points" to the conic concur in the node [1]).

The curve d^4 is a special curve, because its fundamental points coincide with the tangential points D' and D'', so that these are at the same time the points of contact of a double tangent.

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[1]) See my paper "La quartique nodale" (Archives Teyler, t. IX, p. 263).

4. It is easy to find the equation of d^4.

The polar conic of Z with respect to $a_x^3 = 0$ is represented by $a_z a_x^2 = 0$, the polar line by $a_z^2 a_x = 0$. For the tangents out of Z to that conic we have thus

$$a_z a_x^2 b_z^3 = a_z^2 a_x b_z^2 b_x.$$

If these contain the given point Y, then Z is a point of the curve d^4 belonging to Y. So it has, as equation (in current coordinates z):

$$a_y^2 a_z b_z^3 = a_y b_y a_z^2 b_z^2.$$

From this it is again evident, that the polar line of D is double tangent, and that it touches d^4 in the tangential points D' and D''. For, by combination with $a_y^2 a_z = 0$ we find $a_y a_z^2 \,.\, b_y b_z^2 = 0$. The same is obtained by combination with $b_z^3 = 0$; by this is confirmed that d^4 is touched in its points of intersection with the polar conic of D by c^3 and the polar line d.

Out of

$$a_y^2 a_z b_z^3 - a_y b_y a_z^2 b_z^2 = b_y^2 b_z a_z^3 - b_y a_y b_z^2 a_z^2$$

follows that the equation of d^4 can be transformed into

$$\tfrac{1}{2}(a_y a_z b_z^2 - b_y b_z a_z^2)(a_y b_z - a_z b_y) = 0,$$

so also into

$$(a_y b_z - a_z b_y)^2 a_z b_z = 0.$$

Now

$$a_y b_z - a_z b_y = (a_1 b_2)(y_1 z_2) + (a_2 b_3)(y_2 z_3) + (a_3 b_1)(y_3 z_1).$$

If thus we represent the coordinates of the line YZ by ξ_k, the above equation passes into

$$(ab\xi)^2 a_z b_z = 0.$$

This equation expresses that the polar conic of Z is touched by the line YZ [1]).

5. At the same time is evident from this that the line (ξ) cuts its poloconica in two points. This is more closely confirmed by the observation that the poloconica of (ξ) is the locus of the points whose polar conics touch (ξ), from which ensues that it intersects (ξ) in two associated points.

The curve d^4 can therefore be generated by determining the points of intersection of each of the lines s through D with the conjugate poloconica σ. The poloconica describes there a system with index 2. For, when σ passes through any point X the polar conic of X is touched by s. And as two lines s satisfy that condition, X lies on two curves σ. This generation of d^4 with the aid of a system of

[1]) Clebsch, Leçons sur la géométrie, t. II, p. 278.

conics with index 2 and a pencil projective to it is characteristic for the nodal biquadratic curve [1].

6. Each nodal biquadratic curve d^4 of which the nodal tangents pass through the points of contact D' and D'' of a double tangent d is related in the way mentioned above to a c^3.

The polar curve d^3 of D has in D the tangents t' and t'' in common with d^4 and it intersects it in the points of contact R of the six tangents concurring in D. Of the 16 points which d^4 has in common with the system of d^3 and d six lie in D, four in D' and D'', six in the points R. The tangents t' and t'' contain eight of those points; so the remaining eight lie in a conic (curve of BERTINI).

This conic d^2 unites the six points R to the points D' and D''.

Let us now regard the pencil determined by d^4 and the conic d^2 counted twice; one consisting of the double tangent d and a cubic c^3 belongs to it. From this ensues that d^4 is touched by c^3 in the points of contact R of the tangents drawn out of D to d^4.

As d^2 passes through the points R, it is the polar conic of D with respect to c^3; because D' and D'' are the fundamental points, so that DD' and DD'' are touched by d^2 in D' and D'', d is the polar line of D with respect to d^3 and of c^3. So d^4 is the locus of the points associated with respect to c^3 and collinear with D.

7. If d^4 is represented by

$$x_1^2 x_2^2 + x_1 x_2 x_3^2 - c_x^3 x_3 = 0,$$

where

$$c_x^3 = (c_1 x_1 + c_2 x_2)^{(3)}$$

then t', t'', and d are indicated by $x_1 = 0$, $x_2 = 0$, and $x_3 = 0$, and d^3 by $2x_1 x_2 x_3 - c_x^3 = 0$. From

$$(x_1^2 x_2^2 + x_1 x_2 x_3^2 - c_x^3 x_3) - x_3 (2x_1 x_2 x_3 - c_x^3) = 0$$

then follows for d^2 the equation

$$x_1 x_2 - x_3^2 = 0,$$

and from

$$(x_1 x_2 - x_3^2)^2 - (x_1^2 x_2^2 + x_1 x_2 x_3^2 - c_x^3 x_3) = 0$$

we find for c^3

$$c_x^3 - 3x_1 x_2 x_3 + x_3^3 = 0.$$

For the polar conic η^2 of Y with respect to c^3 follows from this

$$c_y c_x^2 - y_1 x_2 x_3 - y_2 x_1 x_3 + y_3 (x_3^2 - x_1 x_2) = 0,$$

for the polar line of D with respect to η^2

$$2y_3 x_3 - y_1 x_2 - y_2 x_1 = 0,$$

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[1]) BOBEK, Denkschriften der Akad. in Wien, Bd. 53, S. 119.

thus for the tangents out of D to the polar conic η^4

$$4y_3\{c_y c_x^2 - y_1 x_2 x_3 - y_2 x_1 x_3 + y_3(x_3^2 - x_1 x_2)\} = (2y_3 x_3 - y_1 x_2 - y_2 x_1)^2.$$

When one of these tangents passes through Y we have

$$4y_3(c_y^3 - 3y_1 y_2 y_3 + y_3^3) = (2y_3^2 - 2y_1 y_2)^2,$$

or

$$y_1^2 y_2^2 + y_1 y_2 y_3^2 - c_y^3 y_3 = 0.$$

From this is again evident that the curve indicated by this equation is the locus of the points associated with respect to c^3 and collinear with D.

This special nodal d^4 is characterized by the property according to which it is touched by a cubic in the six points whose tangents concur in the node. For, when considering the pencil which is determined by d^4 with the conic of BERTINI counted twice it is immediately evident that the remaining points of d^4 lying on this conic are points of contact of a double tangent, which must then also lie on the nodal tangents.

8. We shall now see into what a line (ξ) is transformed by the correspondence of the associated points. To that end we eliminate y_k out of the three equations

$$\xi_y = 0,\ a_y a_x^2 = 0 \text{ and } a_y^2 a_x = 0.$$

Out of the first two we find

$$y_1 : y_2 : y_3 = (a_2 \xi_3) a_x^2 : (a_3 \xi_1) a_x^2 : (a_1 \xi_2) a_x^2.$$

Substitution in the third then produces

$$(ab\xi)(ac\xi)\ a_x b_x^2 c_x^2 = 0.$$

A line ξ is thus transformed into a curve ξ^5 of order five. This could be foreseen, for the two associated points lying on ξ pass in the transformation into each other, whilst the three points of intersection of ξ and c^3 correspond to themselves.

When the point U describes the line ξ, its polar line u envelops the poloconica ξ^2, whilst its polar conic u^2 describes a pencil. From this ensues that ξ^5 is generated by a pencil of conics and a pencil of rays of index 2 projectively related to it. Consequently ξ^5 has nodes in the four basepoints of that pencil and the points associated with U form the pairs of the fundamental involution of pairs appearing on ξ^5 [1]). In connection with this ξ^5 is touched by the poloconica ξ^2 in five points (l. c. p. 48).

[1]) See my paper: "Ueber Curven fünfter Ordnung mit vier Doppelpunkten" (Sitz. Akad. Wien, Bd. 104, S. 47).

Mathematics. — "*On continuous vector distributions on surfaces*". (2nd communication)[1]). By Dr. L. E. J. BROUWER. (Communicated by Prof. D. J. KORTEWEG).

(Communicated in the meeting of February 26, 1910).

§ 1.

The tangent curves to a finite, uniformly continuous vector distribution with a finite[2]) *number of singular points in a singly connected inner domain of a closed curve.*

Let γ be the domain under consideration, then we can represent it on a sphere, so we can immediately formulate on account of the property deduced in the first communication (see there page 855):

THEOREM 1. *A tangent curve, which does not indefinitely approach a point zero, is either a simple closed curve, or its pursuing as well as its recurring branch shows one of the following characters: 1st. stopping at a point of the boundary of γ; 2nd. spirally converging to a simple closed tangent curve; 3rd. entering into a simple closed tangent curve.*

We now shall farther investigate the form (in the sense of analysis situs) of a tangent curve r, of which we assume, that at least *one* of the two branches (e. g. the pursuing branch) approaches indefinitely one or more points zero, i. e. singular points of the vector distribution.

We start the tangent curve in a point A_0 (not a point zero) and we pursue that curve in the following way: By β_ε we understand a distance with the property that in two points lying inside the same geodetic circle described with a radius β_ε, and possessing both a distance $> \varepsilon$ from the points zero, the vectors certainly make an angle $< \frac{1}{8}\pi$ with each other. We farther choose a fundamental series of decreasing quantities $\varepsilon_1, \varepsilon_2, \varepsilon_3, \ldots$ converging to 0, and of corresponding decreasing distances $\beta_{\varepsilon_1}, \beta_{\varepsilon_2}, \ldots$, which all we suppose, if α is the distance of A_0 from the points zero, to be smaller than $\alpha - \varepsilon_1$.

We then prove in the manner indicated in the first communication p. 852, that, when pursuing r from A_0, a point B_0 is reached, possessing a distance β_{ε_1} from A_0; we call the arc A_0B_0 a β_{ε_1}-arc. According to our supposition there now exists a finite number n_1 in

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[1]) For the first communication see these Proceedings Vol. XI 2, p. 850.

[2]) This restriction we shall drop in a following communication.

such a way, that after having completed n_1 β_{ε_1}-arcs, but not yet n_1+1 β_{ε_1}-arcs, we reach a point A_1, where for the first time we have approached the points zero as far as a distance ε_1. Then again there is a finite number n_2 in such a way that, having completed from A_1 n_2, but not yet n_2+1 β_{ε_2}-arcs, we reach a point A_2, where for the first time we have approached the points zero as far as a distance ε_2. From there we pursue r with β_{ε_2}-arcs and continue this process indefinitely.

If we understand by $m(\varepsilon_n)$ the maximum distance from the points zero, which r reaches when being pursued *after* having for the first time approached the points zero as far as a distance ε_n, then a first possibility is, that $m(\varepsilon_n)$ converges with ε_n to zero.

In that case the pursuing branch converges to one single point zero and it is *an arc of simple curve, stopping at that point zero.*

We now suppose the second possibility, that $m(\varepsilon_n)$ surpasses for each ε_n a certain finite quantity e. Then we can effect (by eventually omitting a finite number of terms of the series of ε_n's), that each $\varepsilon_n < \frac{1}{2}e$ and each $\beta_{\varepsilon_n} < \frac{1}{2}e$.

On the pursuing branch then certainly two points P_1 and Q_1 can be indicated both at a distance e from the points zero, and separated on r by at least one point at a distance ε_1 from the points zero, whilst the distance between P_1 and Q_1 is $\leqq \frac{1}{4}\beta_{\varepsilon_1}$. Let P_1S and Q_1U be pursuing β_{ε_1}-arcs, and P_1R and Q_1T recurring β_{ε_1}-arcs.

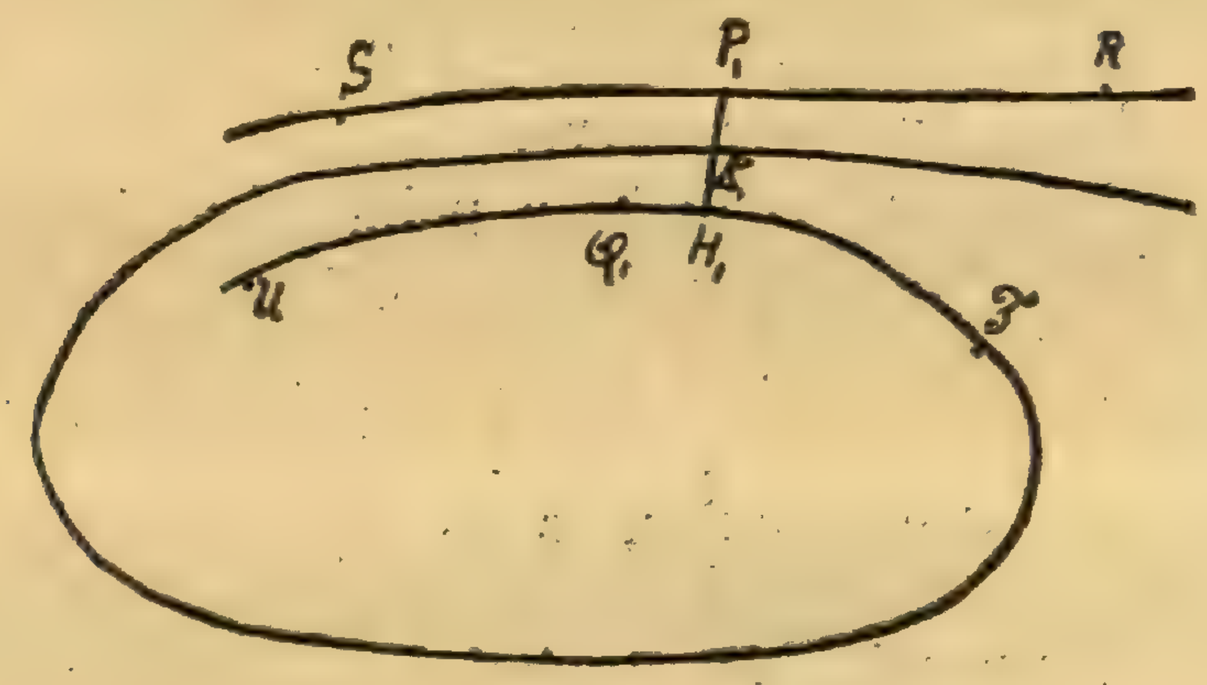


Fig. 1.

Let H_1 be a point of TU, having from P_1 the smallest possible distance, then H_1 cannot coincide with T or U, so that the geodetic arc P_1H_1 is in H_1 normal to the vector direction, and the vector directions in all points of that geodetic arc, forming with

each other an angle $< \frac{1}{8}\pi$, are directed to the same side of the geodetic arc P_1H_1.

Let K_1 be the last point of intersection of the arc P_1H_1 of r with the geodetic arc P_1H_1. Then the arc K_1H_1 of r and the geodetic arc K_1H_1 form a simple closed curve, and we prove in the manner indicated on page 853 of our first communication, that either the pursuing branch of r from H_1 lies in the inner domain, and the recurring branch from K_1 in the outer domain, or the pursuing branch from H_1 in the outer domain, and the recurring branch from K_1 in the inner domain.

Let us first assume that the pursuing branch lies in the *inner domain*, then certainly two points P_2 and Q_2 can be chosen on it, both at a distance e from the points zero and separated on r by at least one point at a distance ε_2 from the points zero, whilst the distance between P_2 and Q_2 is $\leqq \frac{1}{4}\beta_{\varepsilon_2}$. With the aid of those two points we construct in the same way as above now a simple closed curve, consisting of an arc K_2H_2 of r and a geodetic arc K_2H_2, in whose inner domain lies the pursuing branch of r from H_2.

Going on in this way we construct a fundamental series of closed curves $u_1, u_2, u_3, \ldots$ lying inside each other. If there is a domain or set of domains G, common to all the inner domains of these curves (which, as we shall presently show, is really the case) then the boundary of G can only be formed by points belonging to none of the curves $u_1, u_2, u_3, \ldots$ but being limit points of fundamental series of points lying on those curves.

We assume $q > p$, and B to be a point of u_q having a distance $> 3\ \varepsilon_p$ and $> 3\,\beta_{\varepsilon_p}$ from the points zero. Let C be the first point when recurring from B, and D the first point when pursuing from B, which reaches a distance $\frac{1}{2}\beta_{\varepsilon_p}$ from B, then we shall assume for a moment that there exists on u_q, but not on the arc CD, a point S lying at a distance $\leqq \frac{1}{4}\beta_{\varepsilon_p}$ from B, and we shall show that this assumption leads to an absurdity.

Let SV be a recurring $\frac{1}{2}\beta_{\varepsilon_p}$-arc and SW a pursuing $\frac{1}{2}\beta_{\varepsilon_p}$-arc on u_q, then the arcs CD and VW can have no point in common,

and the geodetic arc $K_q H_q$, belonging to u_q, has either no point in common with VW, or none with CD.

In the first case we determine on VW a point M, having from B a distance as small as possible. The geodetic arc BM is then in M normal to VW, and has a last point of intersection N with CD, so that the geodetic arc NM forms with one of the arcs NM of u_q, not containing e.g. the point C, a closed curve; u_q, taken with a certain sense of circuit, would at M enter one of the two domains determined by this closed curve, to leave it no more; further C would lie outside that domain; thus u_q would never be able to reach C, with which the absurdity of our assumption has been proved.

In the second case we determine on CD a point M having from S a distance as small as possible, and on the geodetic arc SM the last point of intersection N with VW. The further reasoning remains analogous to the one just followed: the parts of the arcs VW and CD are only interchanged.

Let now B_ω be the only limit point of a certain fundamental series of points $B_1, B_2, B_3, \ldots$, lying respectively on $u_1, u_2, u_3, \ldots$ We assume that B_ω is not a point zero; it has then for a suitably selected p a distance $> 4\varepsilon_p$ and $> 4\beta_{\varepsilon_p}$ from the points zero.

Let further each m_k be $> p$ and let B_{m_1}, B_{m_2}, $B_{m_3}, \ldots$ be a fundamental series contained in the series just mentioned, whose points have all from B_ω a distance $< \frac{1}{8}\varepsilon_p$ and $< \frac{1}{8}\beta_{\varepsilon_p}$.

If then further on the different u_{m_k} $B_{m_k} D_{m_k}$ are pursuing, $B_{m_k} C_{m_k}$ recurring $\frac{1}{2}\beta_{\varepsilon_p}$-arcs, we prove by the reasoning followed in the first communication p. 854, that there exists a series $C_{n_1} D_{n_1}$, $C_{n_2} D_{n_2}$, $C_{n_3} D_{n_3}, \ldots$ converging uniformly to an arc $C_\omega D_\omega$ of a tangent curve u_ω in such a way, that all arcs $C_{n_k} D_{n_k}$ lie on the same side of $C_\omega D_\omega$.

If we describe round B_ω a geodetic circle with radius $\frac{1}{8}\beta_{\varepsilon_p}$, then it cuts from $C_\omega D_\omega$ an arc FI containing B_ω; this arc divides its inner domain into two regions, into one of which, to be called g, neither the arcs $C_{n_k} D_{n_k}$, nor any other parts of the curves u_{n_k} can penetrate, as they would get there a distance $\leqq \frac{1}{4}\beta_{\varepsilon_p}$ from B_{n_k}.

As further the region g cannot lie outside all curves u_{n_k}, it must lie inside all curves u_{n_k}.

So there is certainly a domain or a set of domains G, common to all the inner domains of the curves u_k, and to the boundary of G belong all points of the limit set λ of the u_k's, which are not points zero, thus also all points of λ, which are points zero, as the latter are limit points of the former ones. So the boundary of G is identical to the limit set of the u_k's, is therefore coherent and identical to its *outer circumference*, whilst abroad from the points zero it consists of tangent curves to the vector distribution, which on account of the existence of the domain g can show nowhere in a non-singular point the character mentioned in theorem 1 sub 3.

We shall now show that a tangent curve r' belonging to the boundary of G cannot have the property of r, that its pursuing or recurring branch converges spirally to the boundary of a domain or set of domains G'.

We should then namely be able to form, in the same way as was done above and in the first communication for r, also for r' a closed curve u'_k consisting of a geodetic arc $\leqq \frac{1}{4}\beta_{\varepsilon_k}$ and an arc φ' of r', joining the same two points K' and H'. And there would exist arcs of r which would converge uniformly to φ' from the same side, e.g. from the inner side of u'_k. But when pursuing such an arc ψ of r situated in sufficient vicinity of φ', we should never be able to return between ψ and φ'.

As farthermore in the case considered here, that the pursuing branch of r lies in the inner domain of u_1, it is also excluded, that r' reaches the boundary of γ, only *one* form remains possible for r', namely *that of an arc of simple curve, starting from a point zero, and stopping at a point zero.* (For the rest these two end points can very well be identical).

Of such tangent curves there can be in the boundary of G at most two, which possess the same end points, when these end points are different; but there can be an infinite number, which are closed in the same point zero. Of these however there are only a finite number, of which the extent surpasses an arbitrarily assumed finite limit. For, each of these contributes to G a domain with an area, which surpasses a certain finite value.

The curves r' whose extent surpasses a certain finite limit are run along by a u_k of sufficient high index in the same order, as they succeed each other on the outer circumference of G. From this ensues *that for all curves r' the pursuing sense belongs to the same sense of circuit of the outer circumference of G.*

If the pursuing branch of r lies in the *outer domain* of u_1, the preceding holds with slight modifications. A point of the limit set of the u_k's now necessarily bounds a region belonging to γ, and lying outside all u_k's, only then when it is not a point of the boundary of γ. The *inner circumference*, to which r *now* converges spirally *on the inner side*, consists here again of arcs of simple curve, which are tangent curves to the vector distribution, but these tangent curves can lie entirely or partially in the boundary of γ.

However they have all again a pursuing sense belonging to the same sense of circuit of the circumference.

We now agree about the following: When a pursuing branch of a tangent curve reaches a point zero, we continue it, if possible, along a pursuing branch, starting from that point zero, and not meeting the former within a certain finite distance; but if such a continuation is impossible, we stop the branch at that point zero, and so we do likewise when the branch has entered into a closed curve or has approximated spirally a circumference. Then we can resume the preceding reasonings as follows:

THEOREM 2. *A tangent curve is either a simple closed curve, or save its ends it is an arc of simple curve, of which the pursuing as well as the recurring branch shows one of the following characters:* 1st. *stopping at a point of the boundary of* γ; 2nd. *stopping at a point zero;* 3rd. *entering into a simple closed tangent curve;* 4th. *spirally converging to a circumference, consisting of one or more simple closed tangent curves.*

From this ensues in particular:

THEOREM 3. *A tangent curve cannot return into indefinite vicinity of one of its points, after having reached a finite distance from it, unless it be to close itself in that point.*

That the last theorem is not a matter of course, is evident from the fact that it does not hold for an annular surface. On this it is easy to construct tangent curves of the form pointed out by LORENTZ (Enz. der Math. Wiss. V 2, p. 120, 121).

We finally notice that the vector distribution considered in this §, does not possess of necessity a singular point (as is the case on the sphere). This is proved directly, by considering in the inner domain of a circle, situated in a Euclidean plane, a vector every where constant.

§ 2.

The structure of the field in the vicinity of a non-singular point.

To classify the singular points we shall surround each of them

with a domain which we shall cover entirely with tangent curves not crossing each other and we shall investigate the different ways in which that covering takes place in different cases. For the sake of more completeness and as an introduction we first do the same for a non-singular point.

Let P be the point under consideration, RS an arc of tangent curve r containing P, UV an arc containing P of an orthogonal curve of the vector distribution. We draw through U and V tangent curves α_0 and α_1, and through R and S orthogonal curves γ and δ, and we let the four points R, S, U, and V converge together to P. Before they have reached P, a moment comes when α_0, α_1, γ, and δ form a curvilinear rectangle, inside which lies P, and inside which lies no point zero of the vector distribution, thus inside which on account of the first communication no closed tangent curve can be drawn.

We shall cover this curvilinear rectangle with tangent curves not crossing each other.

We number α_0 with 0, r with $\frac{1}{2}$, α_1 with 1. Let $Q_{\frac{1}{4}}$ be a point inside or on the rectangle $A_0 B_0 S R$ (fig. 2) having from α_0 and r

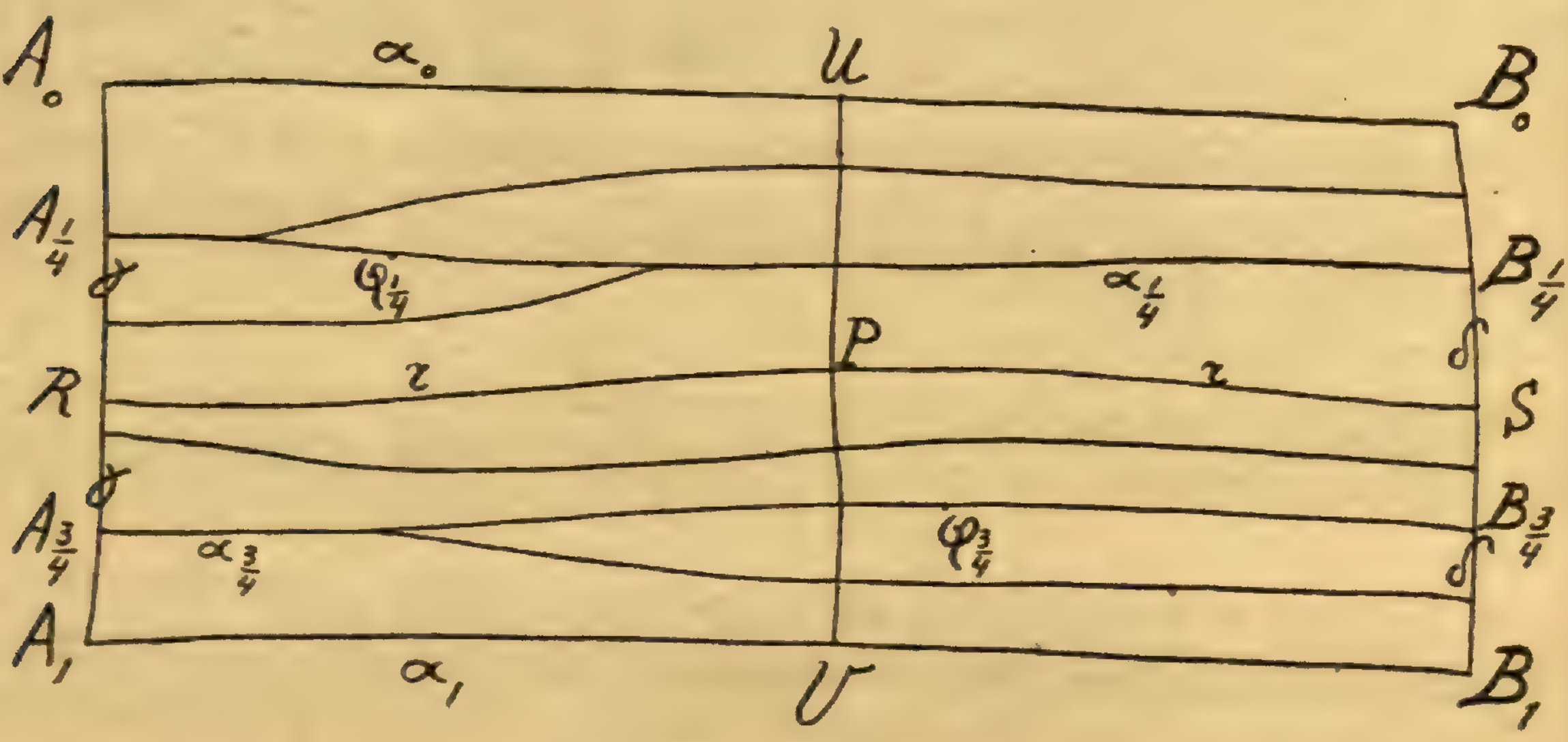


Fig. 2. Non-singular point.

a distance as large as possible. We draw through $Q_{\frac{1}{4}}$ a tangent curve $\alpha_{\frac{1}{4}}$, about which we agree, that, if it meets α_0 or r, we shall continue it, by pursuing or recurring α_0 or r, until we come upon γ or δ.

Then $\alpha_{\frac{1}{4}}$ is a tangent curve joining two points $A_{\frac{1}{4}}$ and $B_{\frac{1}{4}}$ of γ and δ between α_0 and r. In the same way we construct inside

the rectangle $A_1 B_1 S R$ a tangent curve $\alpha_{\frac{3}{4}}$, joining two points $A_{\frac{3}{4}}$ and $B_{\frac{3}{4}}$ of γ and δ between r and α_1. The rectangle $A_0 B_0 B_1 A_1$ is then divided into four regions. In these we choose in the way described above successively the points $Q_{\frac{1}{8}}$, $Q_{\frac{3}{8}}$, $Q_{\frac{5}{8}}$, $Q_{\frac{7}{8}}$, draw through $Q_{\frac{1}{8}}$ a tangent curve $\alpha_{\frac{1}{8}}$ joining two points $A_{\frac{1}{8}}$ and $B_{\frac{1}{8}}$ of γ and δ, and we deal analogously with the other three points.

Going on in this manner we construct for each fraction $\frac{a}{2^n} < 1$ a tangent curve $\alpha_{\frac{a}{2^n}}$ joining two points of γ and δ; two of these curves chosen arbitrarily can coincide *partially*, but they cannot cross each other.

All these tangent curves must now cover everywhere densely the inner domain of the rectangle $A_0 B_0 B_1 A_1$. For, if they left there open a domain G, then a domain G'_n bounded by two tangent curves with indices $\frac{a}{2^n}$ and $\frac{a+1}{2^n}$ would converge to G. For n sufficiently great however the point $Q_{\frac{2a+1}{2^{n+1}}}$ would then lie inside G, thus in contradiction to the supposition also a tangent curve $\alpha_{\frac{2a+1}{2^{n+1}}}$ would pass through G.

From this ensues, that, if we add the limit elements of the tangent curves $\alpha_{\frac{a}{2^i}}$, which are likewise tangent curves, the inner domain of the rectangle $A_0 B_0 B_1 A_1$ is entirely covered, and further there is for each real number between 0 and 1 *one* and not more than *one* of these tangent curves having that number as its index.

§ 3.

The structure of the field in the vicinity of an isolated singular point. First principal case.

We surround the point zero P, supposed isolated, with a simple closed curve c, inside which lies no further point zero. And we assume as a first principal case that c can be chosen in such a way that inside c no simple closed tangent curve exists, inside which P lies. On account of the first communication there can exist inside c neither a simple closed tangent curve, outside which P lies. We now distinguish 2 cases:

a. There exists inside c a simple closed tangent curve ϱ through P. We can then choose c smaller, so that it meets ϱ, thus containing in its inner domain a tangent curve ϱ_1 which (in its pursuing direction) runs from P to c, and another ϱ_2 running from c to P, and we further look for such tangent curves inside c which cross neither ϱ_1 nor ϱ_2. Of the possible kinds of tangent curves mentioned at the conclusion of § 1 we shall agree about those, which enter into a closed tangent curve, to continue them along that tangent curve until they reach either P or c, and to stop there. Spirally converging to an inner circumference cannot appear, as the other end of such a tangent curve would be separated from P as well as from c, and so would determine a closed tangent curve, outside which P would be lying, which is impossible. Neither can appear spirally converging to an outer circumference, as P would have to lie in that outer circumference and the spiral would necessarily have to cross ϱ_1 and ϱ_2.

b. There exists inside c no simple closed tangent curve through P. Then inside c there exists no simple closed tangent curve at all, so that again spirally converging is excluded.

In any case, if we agree not to continue a tangent curve, when it reaches P or c, we can distinguish the tangent curves inside c, and not crossing ϱ_1 and ϱ_2 if the latter exist, into three categories:

1st. *Closed curves, containing P but not reaching c.*

2nd. *Arcs of curve, joining two points of c, but not containing P.*

3rd. *Arcs of curve which run from P to a point of c* (positive curves of the third kind) *or from a point of c to P* (negative curves of the third kind).

Of this third kind there must certainly exist tangent curves. For otherwise the closed sets determined by the curves of the first, and by those of the second kind would cover the whole inner domain of c, thus would certainly possess a point in common; through this point however a curve of the third kind would pass.

So we can commence by constructing one curve of the third kind and we choose eventually ϱ_1 for it. *If possible,* we then draw a second curve of the third kind not crossing the first and we choose eventually ϱ_2 for it. Into each of the two sectors, determined in this way inside c, we introduce if possible again a curve of the third kind, not crossing the already existing ones, and chosen in such a way that it reaches a distance as great as possible from the two curves of the third kind, which bound the sector, whilst, if the new curve terminates somewhere on one of the curves bounding the sector, we further follow the latter curve. In each of the sectors, determined after that in the inner domain of c, we repeat if possible this

insertion, and we continue this process as often as possible, eventually to an indefinite number of insertions.

If in this manner we have obtained an infinite number of tangent curves of the third kind, they determine limit elements which each are either again a tangent curve of the third kind, or contain such a curve as a part. And in particular a fundamental series of positive respectively negative curves of the third kind determines in its limit elements again positive respectively negative curves of the third kind.

After addition of these limit curves of the third kind we are, however, quite sure that no new curves of the third kind not crossing the existing ones can be inserted. This is evident from a reasoning analogous to that followed in § 2. The whole of the curves of the third kind, obtained now, we shall call *a system of base curves of the vicinity of P.*

An arbitrary positive base curve and an arbitrary negative one enclose inside c a sector, of which the area cannot fall below a certain finite limit. For otherwise we should have a fundamental series of positive base curves, and a fundamental series of negative ones, possessing the same base curve as a limit element, which is impossible, as that limit base curve would have to be positive as well as negative.

So the inner domain of c is divided into a finite number of sectors which can be brought under the two following categories:

First category. Sectors bounded by a positive and a negative base curve, between which lie no further base curves. The areas of these sectors surpass a certain finite limit.

Second category. Sectors bounded by two positive (respectively two negative) base curves and containing only positive (respectively negative) base curves. A sector of this category can reduce itself in special cases to a single base curve.

We shall first treat a sector of the *first category* and to that end we first notice that *outside* a curve of the second kind lying in it (i. e. between that curve and c) lie only curves of the second kind, and *inside* a curve of the first kind lying in it only curves of the first kind.

If we draw in the sector a well-ordered series, continued as far as possible, of curves of the second kind enclosing each other, then it converges either to a curve of the second kind, or to two curves of the third kind and between them a finite or denumerable set of curves of the first kind, *not* enclosing each other, and *not* approaching c indefinitely.

If we can construct an infinite number of such series not enclosing

each other, then there are among them which cut from the sector an area as small as one likes, and at the same time the maximum distance, which such a series reaches from c, decreases under each finite limit.

And analogously, if we draw in the sector a well-ordered series, continued as far as possible, of curves of the first kind enclosing each other, it converges either to a curve of the first kind, or to two curves of the third kind and between them a finite or denumerable set of curves of the second kind, *not* enclosing each other, and *not* approaching P indefinitely.

If we can construct an infinite number of such series not enclosing each other, then there are among them which enclose an area as small as one likes, and at the same time the maximum distance, which such a series reaches from P, decreases under each finite limit.

From this ensues that for the sectors of the first category we have to distinguish two cases:

First case. There are curves of the second kind in indefinite vicinity of P. Then the domain of the curves of the second kind is bounded by the two base curves which bound the sector, and a finite or denumerable number of curves of the first kind, *not* enclosing each other, and *not* approaching c indefinitely, in whose inner domains, which we call the *leaves* of the sector, can lie only curves of the first kind.

The region outside the leaves can be covered as follows with curves of the second kind not crossing each other: we first construct one which reaches a distance as great as possible from c and the boundary of the leaves; in this way two new regions are determined, in each of which we repeat this insertion. This process we continue indefinitely, and finally we add the limit curves. That then the region outside the leaves is entirely covered, is evident from the reasoning followed in § 2.

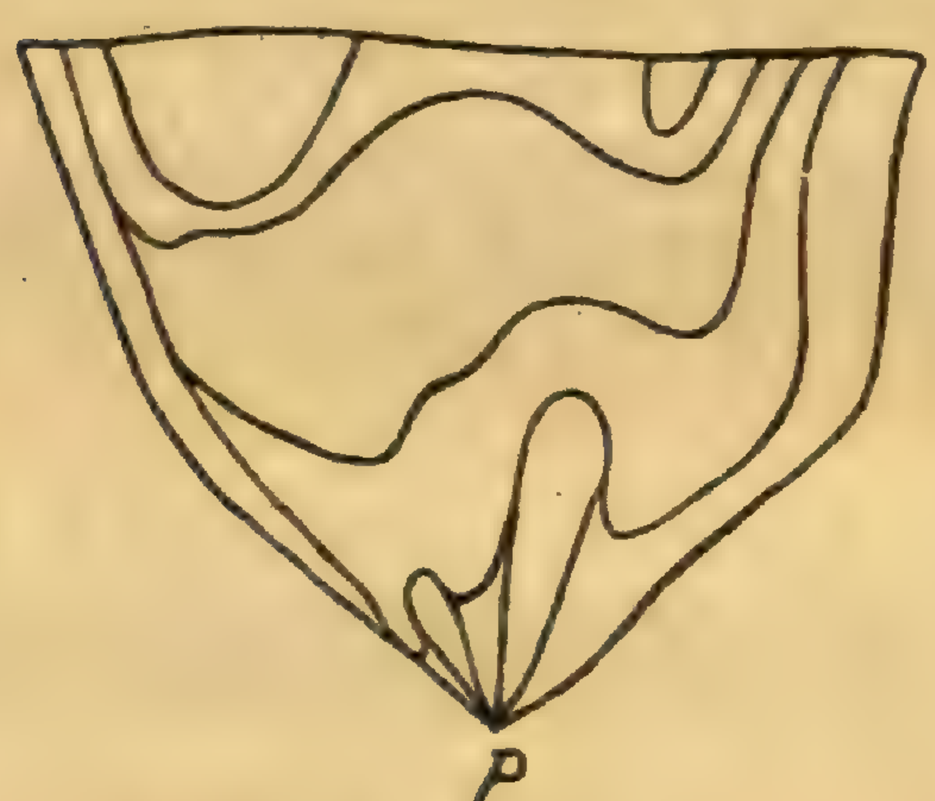


Fig. 3. Hyperbolic sector.

And in the same way we fill each of the leaves with curves of the first kind not crossing each other. The whole of the tangent curves filling the sector finally gets the form indicated in fig. 3. The sectors being in the discussed first case we shall call *hyperbolic sectors*.

Second case. There are no curves of the second kind in indefinite vicinity of P. Then the domains covered by these curves are cut off from

the sector by a finite or denumerable number of curves of the second kind, *not* enclosing each other, and *not* approaching P indefinitely. These domains we take from the sector (consequently modify an arc of c), and there remains a new sector, bounded by the same base curves as the old one, but consisting of one *leaf* inside which lie only curves of the first kind. This leaf we can fill with curves of the first kind not crossing each other (see fig. 4).

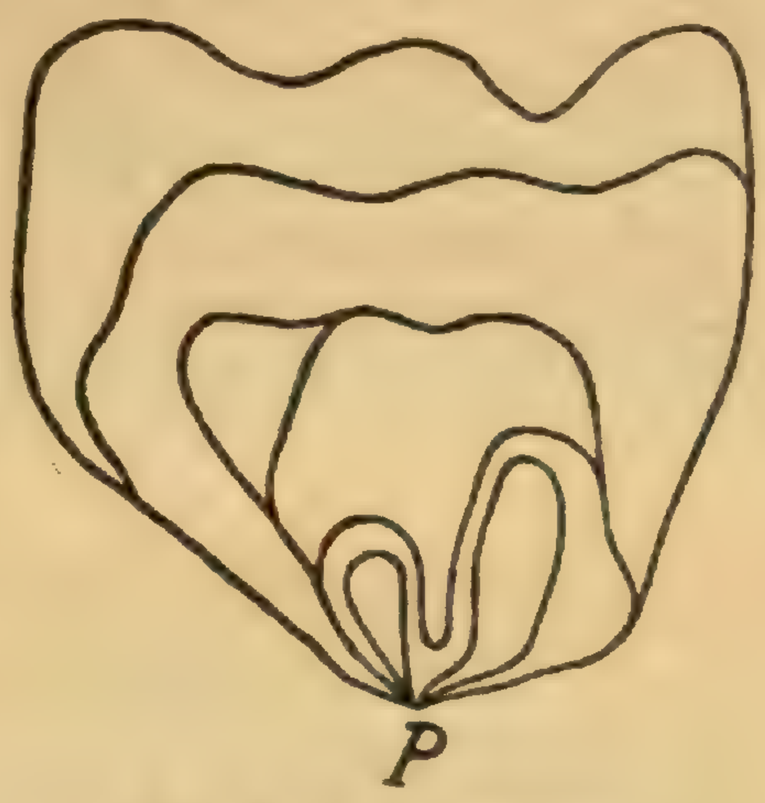


Fig. 4. Elliptic sector.

These sectors of the second case, which are reduced to a single leaf, we shall call *elliptic sectors*.

We now pass to the discussion of a sector of the *second category*, of which, to fix our ideas, we assume, that it is bounded by two positive base curves.

Let us consider the set of points lying in the sector or on its boundary, through which curves of the second kind not crossing the base curves can be drawn. This set of points cannot approach P indefinitely, as otherwise it would give rise to a negative curve of the third kind not crossing the base curves, which is excluded.

In the same way as for the elliptic sectors we destroy the regions covered by this set of points, and there remains a sector of the second category bounded by a modified arc of c, inside which no curves of the second kind not crossing the base curves can be drawn.

In the modified sector we now consider the set of points, through which curves of the first kind not crossing the base curves can be drawn, and it is clear that this set of points cannot indefinitely approach the just now modified curve c. The regions covered by it are therefore bounded by a finite or denumerable number of curves of the first kind, *not* enclosing each other, *not* indefinitely approaching c, and each enclosing a domain which forms a *leaf*, not differing from those appearing in the hyperbolic sectors.

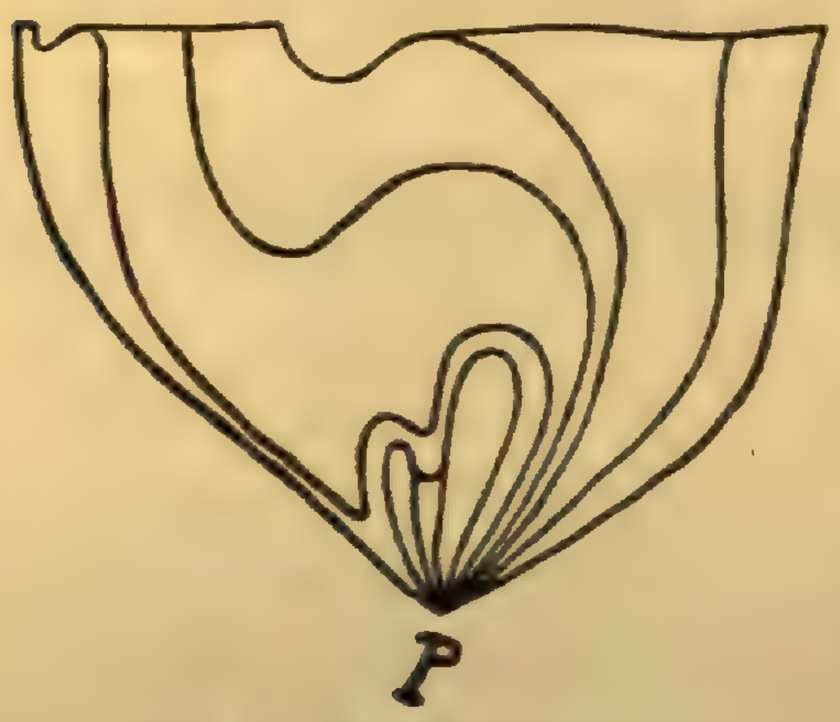


Fig. 5. Parabolic sector.

By the method applied above already several times the region outside the leaves can be filled with curves of the third kind (for instance we can choose for them the system of base curves

present already in the sector), and finally each of the leaves with curves of the first kind (see fig. 5).

The sectors of the second category we shall call *positive* (resp. *negative*) *parabolic sectors.*

In special cases the whole inner domain of c can reduce itself to a single positive (resp. negative) parabolic sector. A point zero where this occurs we shall call a *source point* resp. *vanishing point.*

§ 4.

The structure of the field in the vicinity of an isolated singular point. Second principal case.

In this case any vicinity of P contains a simple closed tangent curve inside which P lies. We can then construct a fundamental series c, c', c'', ... of simple closed tangent curves converging to P, of which each following one lies inside each preceding one, and we can fill in the following way the inner domain of c with tangent curves not crossing each other.

In each annular domain between two curves $c^{(n)}$ and $c^{(n+1)}$ we choose a point having from the boundary of that domain a distance as great as possible and we lay through it a tangent curve situated in the annular domain. According to § 1 it is either closed or it gives rise to two closed curves, situated in the annular domain with its boundary, into which it terminates or to which it converges spirally, and which we draw likewise. (These closed tangent curves can entirely or partially coincide with $c^{(n)}$ or $c^{(n+1)}$). So the annular domain is either made singly connected or it is divided into two or three (annular or singly connected) new domains.

In each of these we again choose a point having from the boundary a distance as great as possible and we lay through it again a tangent curve. A singly connected domain is certainly divided by it into two singly connected domains; on an annular domain it has the effect just now mentioned.

We repeat this process indefinitely. For each domain it can

Fig. 6. Rotation point.

happen only once that it undergoes no division; after that namely it becomes singly connected, so is divided at each new insertion of a tangent curve (see fig. 6).

We finally add the limit curves, and we prove in the same way as in § 2 that then through each point of the inner domain of c passes a tangent curve.

A point zero being in the second principal case we shall call a *rotation point.*

So we can say:

THEOREM 4. *An isolated singular point is either a rotation point, or a vicinity of it can be divided into a finite number of hyperbolic, elliptic, and parabolic sectors.*

The filling of a vicinity of a non-singular point in § 2 furnishes in this terminology two hyperbolic and two parabolic sectors.

We must add the observation that in the most general case, where neither in a singular, nor in a non-singular point the tangent curve is determined, sometimes by a modified method of construction, the structure of the first principal case can be given to a vicinity of a point zero being in the second principal case.

Even the form of the sector division of the first principal case is then not necessarily unequivocally determined. Out of the reasonings of the following § we can, however, deduce that, if modifications are possible in the form of the sector division, the difference of the number of elliptic sectors and the number of hyperbolic sectors always remains the same.

§ 5.

The reduction of an isolated singular point.

For what follows it is desirable to represent the domain γ on a Euclidean plane, and farther to substitute for the curve c a simple closed curve c' emerging nowhere from c, containing likewise P in its inner domain, and consisting of arcs of tangent curves and of orthogonal curves. In the second principal case this is already attained, and in the first principal case we have to modify in a suitable way only those arcs of c which bound the hyperbolic and the parabolic sectors.

In a hyperbolic sector we effect this by choosing a point on each of the two bounding base curves, and by drawing from those points H and K inside into the sector orthogonal arcs not intersecting one another. Then there is certainly an arc of a curve of the second kind

joining a point B of one of these orthogonal arcs with a point C of the other, and we bound the modified sector by the orthogonal arcs HB and CK and the tangent arc BC.

If a parabolic sector is bounded by the base curves k and k', it is always possible to choose between them a finite number of base curves $k_1, k_2, \ldots . k_n$ in such a way, that each k_p and k_{p+1} can be connected, inside the sector but outside the leaves lying in it, by an orthogonal arc. By those orthogonal arcs and the arcs of base curves joining their endpoints we bound the modified sector. The simple closed curve c' obtained in this way has a direction of tangents varying everywhere continuously, with the exception of a finite number of rectangular bends. To a definite sense of circuit of c', which we shall call the positive one, corresponds in each point of c' a definite tangent vector, and for a full circuit of c' that tangent vector describes a positive angle 2π.

We shall now consider two successive parabolic sectors, π_1 and π_2, of which (for the positive sense of circuit) the first is positive, therefore the second negative, and we suppose them to be separated by a hyperbolic sector F. On the orthogonal arcs belonging to the boundary of π_1 the given vector then forms with the tangent vector an angle $\left(2n - \frac{1}{2}\right)\pi$ (measured in the positive sense), on the orthogonal arcs belonging to the boundary of π_2 an angle $\left(2n + \frac{1}{2}\right)\pi$.

The transition takes place along the tangent arc belonging to the boundary of F, by a negative rotation over an angle π of the given vector with respect to the tangent vector.

The same remains the case if we suppose π_1 to be negative, π_2 to be positive.

But if we suppose F to be an elliptic sector, then the transition under discussion takes place along the tangent arc bounding F, by a positive rotation over an angle π of the given vector with respect to the tangent vector.

As now the total angle, which the given vector describes for a full circuit of c', is equal to the total angle which the tangent vector describes plus the total angle which the given vector describes with respect to the tangent vector, the former angle is equal to $\pi\,(2 + n_1 - n_2)$, where n_1 represents the number of elliptic sectors, n_2 the number of hyperbolic ones.

Let further j be an arbitrary simple closed curve enveloping P, but enveloping no other singular point, then we can transform c' into j by continuous modification in such a way, that at every moment

P, but no other singular point, is enveloped by the modified curve. If we consider for each of the intermediary curves the total angle which the given vector describes by a positive circuit, then on one hand it can only have continuous modifications and on the other hand it must remain a multiple of 2π. Thus it remains unchanged, and we can formulate:

THEOREM 5. *The total angle which, by a circuit of a simple closed curve enveloping only one point zero, the vector describes in the sense of that circuit, is equal to* $\pi(2 + n_1 - n_2)$, *where* n_1 *represents the number of elliptic sectors,* n_2 *the number of hyperbolic ones, which appear when a vicinity of the point zero is covered with tangent curves not crossing each other.*

In particular for source points, vanishing points and rotation points this angle is equal to $+ 2\pi$.

We now surround P with a simple closed curve $\varkappa$ which can be supposed as small as one likes, and we leave the vector distribution outside $\varkappa$ and on $\varkappa$ unchanged, but inside $\varkappa$ we construct a modified distribution in the following way:

Let us first suppose that for a positive circuit of $\varkappa$ the vector describes a positive angle $2n\pi$. From an arbitrary point Q inside $\varkappa$ we draw to $\varkappa$ n arcs of simple curve $\beta_1, \beta_2, \ldots \beta_n$, not cutting each other and determining in this order a positive sense of circuit. Let us call ${}_\mu\varkappa$ the arc of $\varkappa$ lying between β_μ and $\beta_{\mu+1}$, and G_μ the domain bounded by β_μ, ${}_\mu\varkappa$ and $\beta_{\mu+1}$. Along β_1 we bring an arbitrary continuous vector distribution becoming nowhere zero and passing on $\varkappa$ into the original one. Then along β_2 such a one passing on $\varkappa$ and in Q into the already existing vectors, that along the boundary of G_1 positively described the vector turns a positive angle 2π. Then along β_3 such a one passing on $\varkappa$ and in Q into the existing vectors, that along the boundary of G_2 positively described the vector turns a positive angle 2π, etc.

As the angle described by the vector in a positive circuit of $\varkappa$ is equal to the sum of the angles described in positive circuits of the boundaries of the domains $G_1, G_2, \ldots G_n$, it is finally evident, that also for a positive circuit of G_n the vector describes a positive angle 2π.

In each of the domains G_μ with boundary $\varkappa_\mu$ we choose a simple closed curve c_μ not meeting $\varkappa_\mu$, of which in a suitable system of coordinates the equation can be written in the form $x_\mu^2 + y_\mu^2 = r^2$.

Inside and on c_μ we introduce a finite continuous vector distribution vanishing only in the point $(o,o)_\mu$, which is directed along the lines

$\frac{y_p}{x_p} = \alpha$ and from the point $(o,o)_p$. This vector describes along c_p a positive angle 2π, just as the existing one along $\varkappa_p$. If then according to SCHOENFLIES we fill the annular domain between $\varkappa_p$ and c_p with simple closed curves enveloping each other and as functions of a cyclic parameter passing continuously into each other, then we can thereby at the same time make the vector distribution along $\varkappa_p$ pass continuously into that along c_p, and in this way give to the annular domain between $\varkappa_p$ and c_p a finite continuous vector distribution vanishing nowhere. Inside $\varkappa_p$ we have now obtained a finite continuous vector distribution, having but *one* point zero, namely the point $(o,o)_p$, and that a source point of very simple structure, which we shall call a *radiating point*.

And the inner domain of $\varkappa$ is covered with a finite continuous vector distribution passing on $\varkappa$ into the original one and possessing inside $\varkappa$, instead of the original point zero P, n radiating points.

Let us furthermore suppose that for a positive circuit of $\varkappa$ the vector describes a negative angle $2n\pi$. In an analogous way as above we then divide the inner domain of $\varkappa$ into n regions G_p with boundaries $\varkappa_p$, and we bring along each of these boundaries such a vector distribution, that for a positive circuit of $\varkappa_p$ the vector describes a negative angle 2π.

The curves c_p are introduced again as above, but inside and on c_p we introduce a finite continuous vector distribution vanishing only in the point $(0,0)_p$, which is directed along the lines $x_p y_p = \alpha$. For a positive circuit this vector describes along c_p a negative angle 2π, just as the existing vector along $\varkappa_p$.

So the annular domain between $\varkappa_p$ and c_p can be filled up in an analogous way as just now with a finite continuous vector distribution vanishing nowhere, and the whole distribution inside $\varkappa_p$ possesses then only *one* point zero, namely the point $(0,0)_p$, having four hyperbolic sectors of very simple form (the four separating parabolic sectors are each reduced to a single line), which structure we characterize by the name of *reflexion point*.

After this the inner domain of $\varkappa$ is covered with a finite continuous vector distribution passing on $\varkappa$ into the original one and possessing inside $\varkappa$, instead of the original point zero P, n reflexion points.

Let us finally suppose that for a circuit of $\varkappa$ the total angle described by the vector is zero. We can then choose inside $\varkappa$ such a simple closed curve c, that in a suitable system of coordinates its equation can be written in the form $x^2 + y^2 = r^2$. Inside and on c we introduce a finite continuous vector distribution vanishing nowhere,

which is directed along the lines $y = a$. The total angle described by this vector along c is zero, just as the one described by the existing vector along $\varkappa$. The annular domain between $\varkappa$ and c can thus be filled up as in the two preceding cases with such a finite continuous vector distribution, that the whole distribution inside $\varkappa$ is now free of points zero.

So we can formulate:

THEOREM 6. *A finite continuous vector distribution with a finite number of points zero can be transformed, by modifications as small as one likes inside vicinities of the points zero which can be chosen as small as one likes, into a new finite continuous vector distribution which has as points zero only a finite number of radiating points, and a finite number of reflexion points.*

In particular those points zero about which the angle, described by the vector for a positive circuit, is positive, are broken up into radiating points; those about which this angle is negative, are broken up into reflexion points; whilst those for which it is zero, vanish.

In a following communication we shall extend this theorem to distributions with an infinite (denumerable or continuous) number of points zero.

§ 6.

Remarks on the tangent curves and singular points on a sphere.

If we have on a sphere a finite continuous vector distribution with a finite number of singular points, then the reasonings of § 1 lead with small modifications to:

THEOREM 7. *A tangent curve to a finite continuous vector distribution with a finite number of singular points on a sphere is either a simple closed curve, or save its ends it is an arc of simple curve, of which the pursuing as well as the recurring branch either stops at a point zero, or enters into a simple closed tangent curve, or converges spirally to a circumference consisting of one or more simple closed tangent curves.*

From this ensues that also on a sphere a tangent curve cannot return into indefinite vicinity of one of its points, after having reached a finite distance from it, unless it be, to close itself in that point.

Out of the reasoning of § 1 we can deduce farthermore without difficulty that a fundamental series of closed tangent curves with the property that of the two domains determined by one of them,

one contains no points of the preceding, the other no points of the following curves, converges either to a single singular point, or to the outer circumference, consisting of simple closed tangent curves, of a domain or set of domains.

Let now an arbitrary finite continuous vector distributionon a sphere be given. On account of § 5 we reduce it by means of indefinitely small modifications to a "reduced distribution", possessing as singular points only radiating points and reflexion points, and we investigate the tangent curves of that reduced distribution.

A closed tangent curve can possess no radiating points, but reflexion points it can possess (its tangent direction shows there a rectangular bend).

On the other hand a tangent curve can only stop at a radiating point.

We now consider an arbitrary tangent curve: according to theorem 7 it is either an arc of simple curve joining two radiating points, or it gives rise to a simple closed tangent curve j_0, which divides the sphere into two domains G and G'.

Then on j_0 no radiating point can lie, but we shall prove, that in G as well as in G' there must lie one.

If namely there were no radiating point in G, we could consider within G a new tangent curve, and as this would not be able to stop in G, it would on account of theorem 2 give rise to a new simple closed tangent curve j_1 enclosing a domain G_1 being a part of G. Within G_1 we could again consider an arbitrary tangent curve, and in this way we should arrive at a simple closed tangent curve j_2 enclosing a domain G_2 being a part of G_1.

Continuing this process indefinitely we construct a fundamental series of closed tangent curves $j_0, j_1, j_2, j_3, \ldots$, which cannot converge to a single singular point, as neither a radiating point nor a reflexion point contains closed tangent curves in an indefinitely small vicinity. On account of the remark made at the beginning of this § there must thus be at least *one* domain G_ω, bounded by a simple closed tangent curve j_ω, and contained in each of the domains $G_1, G_2, G_3, \ldots$.

Within G_ω we could again construct a closed tangent curve $j_{\omega+1}$ bounding a domain $G_{\omega+1}$ being a part of G_ω, and we could continue this process to *any* index of the second class of numbers, which on the other hand is impossible, as the set of domains $G - G_1$, $G_1 - G_2, \ldots G_\omega - G_{\omega+1}, \ldots G_\alpha - G_{\alpha+1}, \ldots$ must remain denumerable.

So we finally formulate:

THEOREM 8. *A reduced distribution on a sphere possesses at least two radiating points.*

Fig. 1.

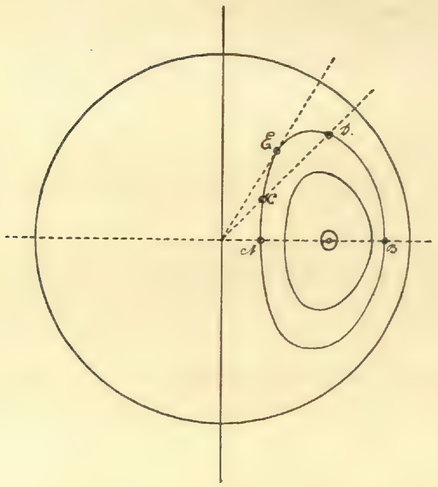


Fig. 2.

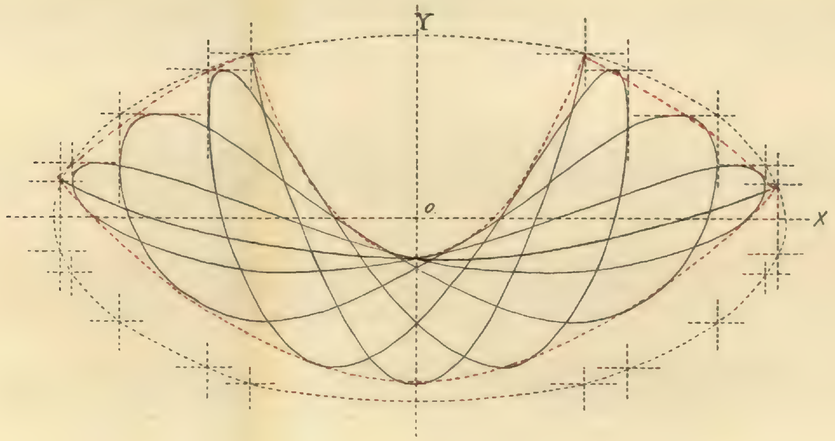


Fig. 3.

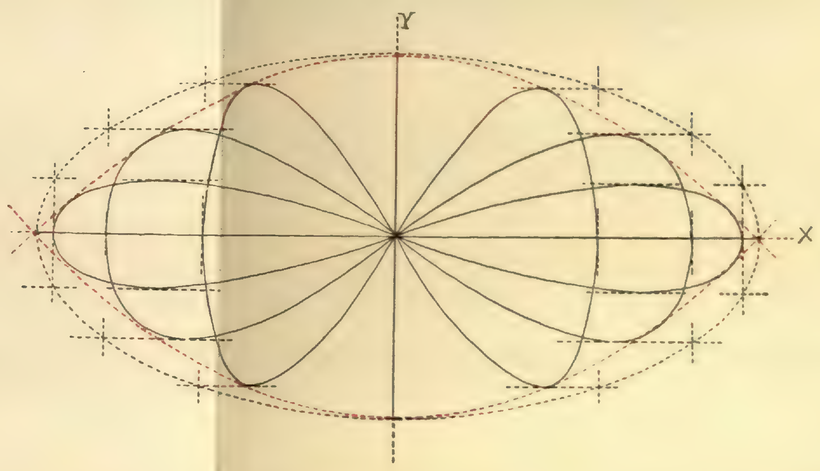


Fig. 4.

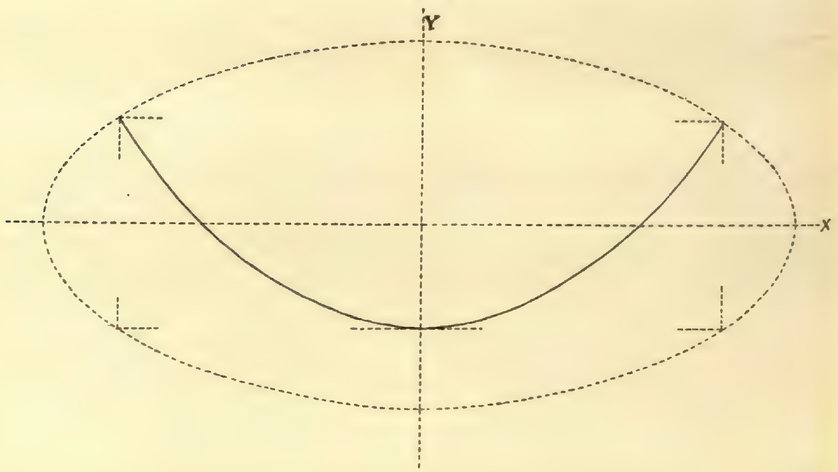


Fig. 5.

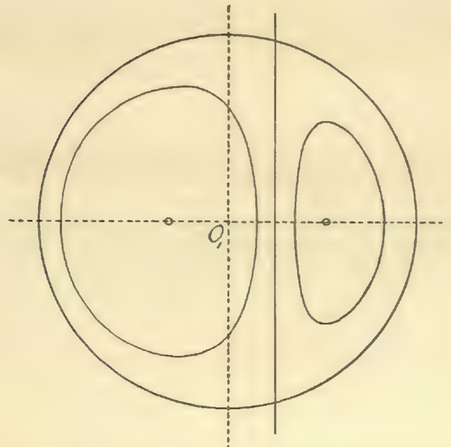


Fig. 6a.

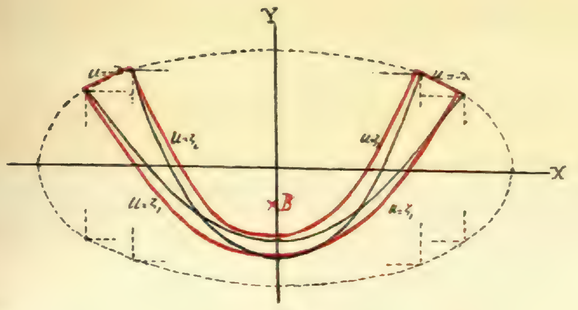


Fig. 6b.

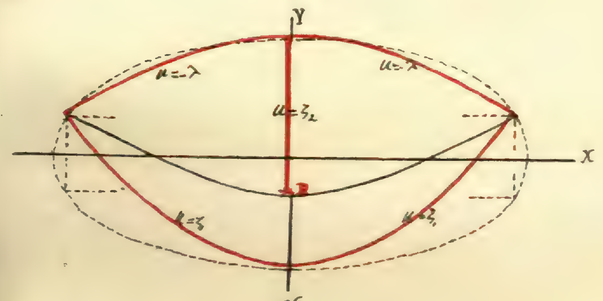


Fig. 6c.

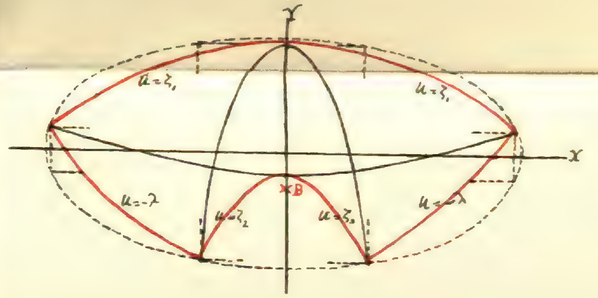
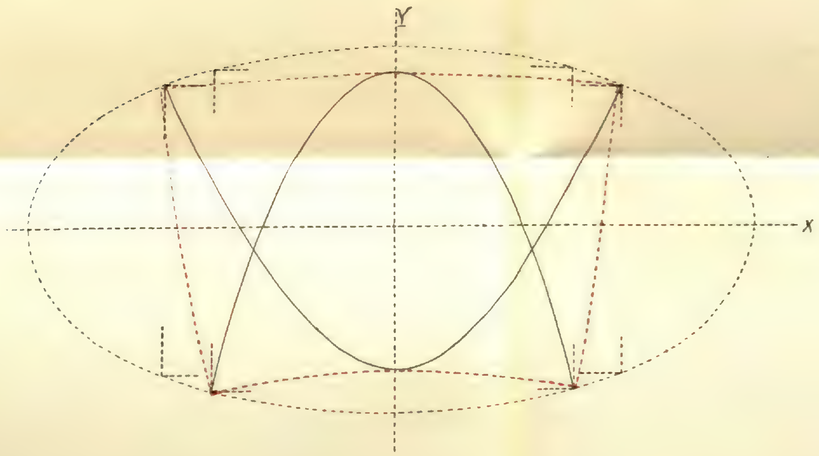


Fig. 7.



Mathematics, — "*The oscillations about a position of equilibrium where a simple linear relation exists between the frequencies of the principal vibrations.*" (Second part). By H. J. E. BETH. (Communicated by Prof. D. J. KORTEWEG.)

(Communicated in the meeting of February 26, 1910).

$S = 4$. [1])

§ 14. In this case the ordinary expansions in series hold as long as $\frac{\varrho}{n_1}$ is great with respect to $\left(\frac{h}{R_1}\right)^2$ (see page 7 of the paper by Prof. KORTEWEG, mentioned above). The difficulty arises as soon as $\frac{\varrho}{n_1}$ has fallen to the order $\left(\frac{h}{R_1}\right)^2$. The calculations not getting simpler with the absence of a residue of relation, we shall immediately assume a residue of relation of order h^2.

When the relation

$$n_2 + \varrho = 3n_1$$

exists and we proceed to investigate with a view to this which terms in (2) (page 620 of these Proceedings) become disturbing in the sense indicated in § 3, we easily see that no terms of order h^2 appear among the disturbing ones. So when determining the first approximation we may omit the terms of order h^3 in the equation of the surface, which terms agree with the just mentioned terms of order h^2. It then becomes

$$z = \frac{1}{g}(c_1x^2 + c_2y^2 + e_1x^4 + e_2x^3y + e_3x^2y^2 + e_4xy^3 + e_5y^4);$$

for we need not take for the first approximation in the equations of movement any terms of higher order than h^3.

The abridged equations of motion, containing only terms of order h, still run as follows:

$$\left.\begin{aligned} \ddot{x} + 2c_1\, x &= 0, \\ \ddot{y} + 2c_2\, y &= 0. \end{aligned}\right\}$$

Now

$$n_1 = \sqrt{2c_1} \quad , \quad n_2 = \sqrt{2c_2}$$

are the frequencies of the principal vibrations.

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[1]) For the case $S = 3$ see 1st part, pages 619—635 of these Proceedings.

So

$$2c_3 = (3n_1 - \varrho)^2.$$

We change the abridged equations into:

$$\left.\begin{aligned} \ddot{x} + n_1^2\, x &= 0, \\ \ddot{y} + 9n_1^2\, y &= 0\,; \end{aligned}\right\}$$

but then we must admit into the function R a term:

$$3n_1\, \varrho\, y^2.$$

The canonical solution of the abridged equations is:

$$x = \frac{\sqrt{\alpha_1}}{n_1} \cos(n_1 t + 2n_1\beta_1),$$

$$y = \frac{\sqrt{\alpha_2}}{3n_1} \cos(3n_1 t + 6n_1\beta_2).$$

To find which functions the α's and β's are of t, we must investigate which form the function R now assumes.

§ 15. As the disturbing terms in the equations of motion are of order h^3 we shall find that $\dot{\alpha}_1$, $\dot{\alpha}_2$, $\dot{\beta}_1$, and $\dot{\beta}_2$ can never exceed order h. Of this we may make use to simplify the terms of order h^3 containing x, y, x^2, and y^2. We may namely replace in those terms:

$\dot{x}^2$ by $\alpha_1 - n_1^2\, x^2$,

$\dot{y}^2$ „ $\alpha_2 - 9n_1^2\, y^2$,

$\ddot{x}$ „ $-n_1^2\, x$

and

$\ddot{y}$ „ $-9n_1^2\, y$.

Then the equations become:

$$\ddot{x} + n_1^2\, x + 4e_1\, x^3 + 3e_2\, x^2 y + 2e_3\, xy^2 + e_4\, y^3 + \\ + \frac{n_1^4}{g^2}(\alpha_1 + 9\alpha_2)\, x - \frac{2n_1^6}{g^2}(x^2 + 81\, y^2)\, x = 0.$$

$$\ddot{y} + 9n_1^2\, y - 6n_1\, \varrho y + e_2\, x^3 + 2e_3\, x^2 y + 3e_4\, xy^2 + 4e_5\, y^3 + \\ + \frac{9n_1^4}{g^2}(\alpha_1 + 9\alpha_2) y - \frac{18n_1^6}{g^2}(x^2 + 81\, y^2)\, y = 0.$$

Now the terms of order h^3 are all disturbing except $e_4 y^3$ in the first and $3e_4\, xy^2$ in the second equation; so these may be omitted.

The terms $3e_2\, x^2 y$ in the first and $e_2\, x^3$ in the second equation owe their disturbing property to the supposed relation.

The remaining terms are always disturbing, also when no relation exists.

To transform the equations to such a form that the disturbing terms may be regarded as derivatives of one and the same function resp. to x and y, let us consider the term with xy^2 in the first and that with x^2y in the second equation. If we substitute the expressions found above as first approximation for x and y in these terms, after the development of the products and powers of the cosines among others terms will appear, differing only in coefficient from the expressions indicated for x and y; the remaining terms which appear are not disturbing. From this ensues that we may replace:

$$\text{in the first equation: } xy^2 \text{ by } \frac{1}{18}\frac{a_2}{{n_1}^2}x,$$

$$\text{in the second equation: } x^2y \text{ by } \frac{1}{2}\frac{a_1}{{n_1}^2}y.$$

Accordingly the equations may be written:

$$\left.\begin{aligned} &\ddot{x} + {n_1}^2 x + \left(4e_1 - \frac{2{n_1}^6}{g^2}\right)x^3 + 3e_2 x^2 y + \left(\frac{e_2}{9{n_1}^2}a_2 + \frac{{n_1}^4}{g^2}a_1\right)x = 0,\\ &\ddot{y} + 9{n_1}^2 y + \left(4e_5 - \frac{1458{n_1}^6}{g^2}\right)y^3 + e_2 x^3 + \\ &\qquad + \left(-6n_1\varphi + \frac{e_2}{{n_1}^2}a_1 + \frac{81{n_1}^4}{g^2}a_2\right)y = 0. \end{aligned}\right\}$$

We thus see that they take the form of:

$$\left.\begin{aligned} &\ddot{x} + {n_1}^2 x - \frac{\partial R}{\partial x} = 0,\\ &\ddot{y} + 9{n_1}^2 y - \frac{\partial R}{\partial y} = 0; \end{aligned}\right\}$$

where:

$$-R \equiv \left(e_1 - \frac{{n_1}^6}{2g^2}\right)x^4 + \left(e_5 - \frac{729{n_1}^6}{2g^2}\right)y^4 + \frac{1}{2}\left(\frac{e_2}{9{n_1}^2}a_2 + \frac{{n_1}^4}{g^2}a_1\right)x^2 + \\ + \frac{1}{2}\left(-6n_1\varphi + \frac{e_2}{{n_1}^2}a_1 + \frac{81{n_1}^4}{g^2}a_2\right)y^2 + e_2 x^3 y.$$

§ 16. We must now write R as function of the α's and β's by substituting for x and y, in the expressions obtained, the expressions by which they are represented at first approximation, and by retaining only those terms in which t does not appear explicitly. Thus we arrive at:

$$-R = \frac{1}{2}aa_1^2 + ba_1a_2 + \frac{1}{2}ca_2^2 + \varphi' h^2 a_2 + m_1 a_1^{\frac{3}{2}} a_2^{\frac{1}{2}} \cos\varphi,$$

where:

$$a = \frac{3e_1}{4n_1^4} + \frac{n_1^2}{8g^2},$$

$$b = \frac{e_3}{18n_1^4},$$

$$c = \frac{e_5}{108n_1^4} + \frac{9n_1^2}{8g^2},$$

$$m_1 = \frac{e_3}{24n_1^4},$$

$$\varrho = -\frac{\varrho}{6n_1h^2},$$

$$\varphi = 6n_1(\beta_1 - \beta_2).$$

The system of equations giving the time-variability of the α's and β's, is now:

$$\left.\begin{aligned}
\frac{d\alpha_1}{dt} &= 2Nm_1\,\alpha_1^{\frac{3}{2}}\,\alpha_2^{\frac{1}{2}}\sin\varphi,\\
\frac{d\alpha_2}{dt} &= -2Nm_1\,\alpha_1^{\frac{3}{2}}\,\alpha_2^{\frac{1}{2}}\sin\varphi,\\
\frac{d\beta_1}{dt} &= a\alpha_1 + b\alpha_2 + \frac{3}{2}m_1\,\alpha_1^{\frac{1}{2}}\,\alpha_2^{\frac{1}{2}}\cos\varphi,\\
\frac{d\beta_2}{dt} &= b\alpha_1 + c\alpha_2 + \varrho' h^2 + \frac{1}{2}m_1\,\alpha_1^{\frac{3}{2}}\,\alpha_2^{-\frac{1}{2}}\cos\varphi;
\end{aligned}\right\} \quad . \quad (17)$$

where N is put instead of $3n_1$.

From this system it appears at once that:

$$\frac{d\alpha_1}{dt} + \frac{d\alpha_2}{dt} = 0,$$

therefore

$$\alpha_1 + \alpha_2 = constant.$$

So we put:

$$\alpha_1 = R_0^2\,n_1^2\,h^2\,\zeta \quad , \quad \alpha_2 = R_0^2\,n_1^2\,h^2\,(1-\zeta).$$

Furthermore according to § 4:

$$\frac{1}{2}a\alpha_1^2 + b\alpha_1\,\alpha_2 + \frac{1}{2}c\alpha_2^2 + \varrho' h^2\,\alpha_2 + m_1\,\alpha_1^{\frac{3}{2}}\,\alpha_2^{\frac{1}{2}}\cos\varphi = constant$$

is an integral of the system.

By introduction of ζ this integral takes the form of:

$$\zeta\sqrt{\zeta(1-\zeta)}\cos\varphi = p\zeta^2 + q\zeta + r, \quad . \quad . \quad . \quad . \quad . \quad (18)$$

where:

$$p = \frac{1}{m_1}\left(-\frac{1}{2}a + b - \frac{1}{2}c\right),$$

$$q = \frac{1}{m_1}\left(-b + c + \frac{\varrho'}{n_1^2 R_0^2}\right),$$

$$r = \frac{1}{m_1}\left(-\frac{1}{2}c - \frac{\varrho'}{n_1^2 R_0^2} + \frac{C}{n_1^4 R_0^4}\right),$$

where C represents a constant, dependent on the initial state.

The first equation of (17) becomes by the introduction of ζ:

$$\frac{d\zeta}{dt} = \frac{2}{9} m_1 R_0^2 N^3 h^3 . \zeta \sqrt{\zeta(1-\zeta)} . \sin\varphi. \quad . \quad . \quad . \quad . \quad (19)$$

By eliminating φ between (18) and (19) we arrive at:

$$\frac{d\zeta}{\sqrt{\zeta^3(1-\zeta) - (p\zeta^2 + q\zeta + r)^2}} = \pm \frac{2}{9} m_1 R_0^2 N^3 h^2 . dt.$$

Let

$$f(\zeta) \equiv \zeta^3(1-\zeta) - (p\zeta^2 + q\zeta + r)^2,$$

then $f(\zeta) > 0$ for the initial value of ζ, but $f(\zeta) < 0$ for $\zeta = 0$ and $\zeta = 1$; so $f(\zeta)$ becomes zero for two values ζ_1 and ζ_2 lying between 0 and 1.

So ζ will generally vary periodically between two limits. It may be expressed in the time with the aid of elliptic functions, after which β_1, β_2, x, and y are also known as functions of the time.

For the extreme values *zero* and *one* of the modulus $\varkappa$ of the elliptic functions ($\varkappa = \sqrt{\frac{(\beta-\alpha)(\zeta_2-\zeta_1)}{(\beta-\zeta_2)(\alpha-\zeta_1)}}$, when the equation $f(\zeta) = 0$ has two real roots α and β besides ζ_1 and ζ_2) we get special cases.

Osculating curves.

§ 17. At first approximation we have found:

$$x = \frac{\sqrt{a_1}}{n_1} \cos(n_1 t + 2n_1 \beta_1),$$

$$y = \frac{\sqrt{a_2}}{3n_1} \cos(3n_1 t + 6n_1 \beta_2),$$

where the α's and β's slowly vary with the time.

By introduction of ζ and φ and by change of the origin of time we find that we may determine the equation of an osculating curve by eliminating t between

$$x = R_0\, h \sqrt{\zeta}\, \cos n_1\, t$$

and

$$y = \frac{1}{3} R_0\, h \sqrt{1-\zeta}\, \cos (3 n_1\, t - \varphi).$$

For ζ and φ we must substitute the values, which these quantities have at the moment for which we wish to know the osculating curve.

The osculating curves are Lissajous curves answering to the value $\frac{1}{3}$ for the ratio of the periods of the vibrations. They are described in the rectangles having as sides $2\ R_0\, h \sqrt{\zeta}$ and $\frac{2}{3} R_0\, h \sqrt{1-\zeta}$. As ζ varies between two limits the rectangles in which the curves are described lie between two extremes. The vertices lie on the circumference of an ellipse having $2\, R_0\, h$ and $\frac{2}{3} R_0\, h$ as lengths of axes.

The shape of the curve described in a definite rectangle is still dependent on the value of φ, i. e. on the value of the difference in phase at the moment of the greatest deviation to the right.

To an arbitrary value of φ the wellknown Lissajous curve with two nodes of fig. 8 answers. For $\varphi = \frac{\pi}{2}$ or $\frac{3\pi}{2}$ the curve is symmetrical in respect to the axes; the nodes lie in the X-axis on either side of O at distances $\frac{1}{2} R_0\, h$ (fig. 9). For $\varphi = 0$ or π we get a curve, which is described in both directions alternately and which passes through O (fig. 10).

In fig. 11 we find some of those osculating curves represented for a definite case of motion: *two* belonging to $\varphi = \pi$; *two* for $\varphi = \frac{\pi}{2}$, and *one* for an arbitrary value of $\varphi \left(> \frac{\pi}{2} \right)$.

Out of (19) follows that $\frac{d\zeta}{dt} = 0$ for $\sin \varphi = 0$. In the extreme rectangles the curves are described which we have for $\varphi = 0$ or π. Now a number of different cases are possible, of which we get a clear representation by representing equation (18) in polar coordinates. In fig. 12 some of the curves obtained in this way are represented, where φ is taken as polar angle, $\sqrt{1-\zeta}$ as radius vector. The different shapes of the curves correspond to the roots of the equation:

$$\zeta^3 (1 - \zeta) - (p\zeta^2 + q\zeta + r)^2 = 0. \quad . \quad . \quad . \quad . \quad (20)$$

The cases are:

1. The curve indicated in the figure by --- keeps to the right or to the left of O_1; φ changes between two limits; the limits are equal and opposite; the positive is smaller than $\frac{\pi}{2}$. For the extreme values of ζ we find φ either both times 0 or both times π.

2. The curve — — — intersects the straight line $\varphi = \frac{\pi}{2}$ at two points above O_1 and at 2 points below O_1. For the extreme values of ζ we again find φ either both times 0 or both times π.

3. The curve consists of two closed parts (a continuous line in the figure), which surround O_1. Now φ assumes all values. For the extreme values of ζ $\varphi = 0$ *one* time $\varphi = 0$ and $\varphi = \pi$ the other.

The transition case between 2 and 3 is represented by — . — . —.

Fig. 11 relates to the 2nd case; for the two extreme values of ζ we find $\varphi = \pi$.

Special cases.

§ 18. These occur for the extreme values of the modulus $\varkappa$ of the elliptic functions; two roots of equation (20) have coincided.

1. $\varkappa = 1$. The elliptic functions pass into hyperbolic ones. The geometrical representation just now discussed of the relation between ζ and φ and already mentioned as transition case between the second and third cases has a node situated on the axis of the angles. The form of motion approaches asymptotically to a form of motion belonging to $\varphi = 0$ or $\varphi = \pi$.

2. $\varkappa = 0$. The elliptic functions pass into goniometrical ones. The curve of fig. 12 becomes an isolated point C (special case belonging to the 1st case of § 17 as limiting case) or it consists of an isolated point and a closed curve (special case belonging to the 3rd case of § 17 as limiting case). If the initial value of ζ coincides with the twofold root of (20) we find that ζ remains constant; φ is continually 0 or π. Thus the same curve is continually described.

Arbitrary mechanism with 2 degrees of freedom for which $S = 4$.

§ 19. In the case that $n_2 = 3n_1 + \varrho$ the terms of order h^3 can give no disturbing terms in the equations of motion.

So we may write:

$$U = \tfrac{1}{2} n_1^2 q_1^2 + \tfrac{1}{2} n_2^2 q_2^2 + U_4 ,$$

where U_4 represents a homogeneous function of degree 4 in q_1 and q_2. Furthermore we find:

$$T = \tfrac{1}{2}\dot{q}_1^2 + \tfrac{1}{2}\dot{q}_2^2 + \tfrac{1}{2}P_1\dot{q}_1^2 + P_2\dot{q}_1\dot{q}_2 + \tfrac{1}{2}P_3\dot{q}_2^2;$$

where:

$$P_1 = a_1 q_1^2 + a_2 q_1 q_2 + a_3 q_2^2,$$
$$P_2 = b_1 q_1^2 + b_2 q_1 q_2 + b_3 q_2^2,$$
$$P_3 = c_1 q_1^2 + c_2 q_1 q_2 + c_3 q_2^2,$$

the a's, b's, and c's being constants.

The equations of LAGRANGE become:

$$\left.\begin{aligned}
\ddot{q}_1 + n_1^2 q_1 = &- P_1\ddot{q}_1 - P_2\ddot{q}_2 - \frac{1}{2}\frac{\partial P_1}{\partial q_1}\dot{q}_1^2 - \frac{\partial P_1}{\partial q_2}\dot{q}_1\dot{q}_2 + \\
&+ \left(\frac{1}{2}\frac{\partial P_2}{\partial q_1} - \frac{\partial P_2}{\partial q_2}\right)\dot{q}_2^2 - \frac{\partial U_4}{\partial q_1}. \\
\ddot{q}_2 + n_2^2 q_2 = &- P_2\ddot{q}_1 - P_3\ddot{q}_2 + \\
&+ \left(\frac{1}{2}\frac{\partial P_1}{\partial q_2} - \frac{\partial P_2}{\partial q_1}\right)\dot{q}_1^2 - \frac{\partial P_2}{\partial q_1}\dot{q}_1\dot{q}_2 - \frac{1}{2}\frac{\partial P_3}{\partial q_2}\dot{q}_2^2 - \frac{\partial U_4}{\partial q_2}.
\end{aligned}\right\}$$

In the same way as was done in § 15 we may replace $\ddot{q}_1$, $\ddot{q}_2$, q_1^2, and q_2^2 in the terms of order h^3 by others.

Now in the first equation a term $-a_2 q_1 \dot{q}_1 \dot{q}_2$ appears which we must consider separately (in the second equation also there are terms containing $q_1 q_2$, but these are not disturbing).

We introduce for this a new variable q'_1 in such a way that:

$$q'_1 = q_1 + \frac{1}{4}a_2 q_1^2 q_2.$$

Then we find:

$$\ddot{q}'_1 = \ddot{q}_1 + \frac{1}{4}a_2 q_1^2\ddot{q}_2 + \frac{1}{2}a_2 q_2(q_1\ddot{q}_1 + \dot{q}_1^2) + a_2 q_1\dot{q}_1\dot{q}_2,$$

where $\ddot{q}_1$ and $\ddot{q}_2$ in the terms of order h^3 may again be simplified.

Of the terms now appearing in the equations of motion the following are disturbing: in the first equation those with $h^2 q_1$, q_1^3, $q_1^2 q_2$ and $q_1 q_2^2$, in the second those with $h^2 q_2$, q_1^3, q_2^3 and $q_1^2 q_2$. Now just as in § 15 the terms with $q_1 q_2^2$ in the first equation, those with $q_1^2 q_2$ in the second equation may still be simplified.

If we perform these calculations the result proves that the terms of order h^3 to be inserted in the equations may be put in this form:

$Ph^2 q_1 + eq_1^2 q_2 + cq_1^3$ in the first equation.

$Qh^2 q_2 + fq_1^3 + dq_2^3$ „ „ second „

Here P and Q are homogeneous quadratic functions of $\sqrt{\alpha_1}$ and $\sqrt{\alpha_2}$; and

$$e = \frac{1}{4}(3n_1^2 - n_2^2)\,a_2 + b_1 n_2^2 - 3l,$$

$$f = -\frac{1}{2}n_1^2 a_2 + 3b_1 n_1^2 - l.$$

(The terms $-3l$ in e and $-l$ in f originate from the term $lq_1^3 q_2$ appearing in U_4).

In the terms of higher order we may substitute $3n_1$ for n_2 in the coefficients. We then find:

$$e = 3\left\{\left(-\frac{1}{2}a_2 + 3b_1\right)n_1^2 - l\right\},$$

$$f = \left(-\frac{1}{2}a_2 + 3b_1\right)n_1^2 - l.$$

So we find that

$$e = 3f.$$

We may now write the equations of motion:

$$\left.\begin{aligned} \ddot{q}_1 + n_1^2 q_1 &= \frac{\partial R}{\partial q_1}, \\ \ddot{q}_2 + n_2^2 q_2 &= \frac{\partial R}{\partial q_2}; \end{aligned}\right\}$$

where

$$R = \frac{1}{2}Ph^2 q_1^2 + \frac{1}{2}Qh^2 q_2^2 + fq_1^3 q_2 + \frac{1}{4}cq_1^4 + \frac{1}{4}dq_2^4.$$

So they get the same form as for the simple mechanism so that in case $S = 4$ also the horizontal projection of the point moving over the surface may be regarded as representative point for an arbitrary mechanism with 2 degrees of freedom.

$S = 2$.

§ 20. So we suppose that the relation exists:

$$n_1 = n_2 + \varrho,$$

where $\frac{\varrho}{n_1}$ is of order $\left(\frac{h}{R_1}\right)^2$. However, as we have already seen in the cases $S = 3$ and $S = 4$ in which way such a residue of relation may be taken into account by inserting in the function R a term with $\varrho\alpha_2$, we restrict ourselves here to the case that the residue of relation is zero, therefore:

$$n_2 = n_1 = n.$$

For the surface the lowest point is an umbilical point. To this belongs as special case the surface of revolution with the Z-axis as axis of revolution, which case is treated by Prof. KORTEWEG at the close of his treatise quoted before.

Omitting the terms of higher order than h^4, because in the equations of motion we admit no terms of higher order than h^3, and omitting the terms of order h^3, because in the equations of motion no terms of order h^2 can be disturbing, we may write the equation of the surface:

$$z = \frac{1}{g}(c_1x^2 + c_2y^2 + e_1x^4 + e_2x^3y + e_3x^2y^2 + e_4xy^3 + e_5y^4),$$

where we avail ourselves of the fact, that by means of a rotation of the system of coordinates round the Z-axis the coefficients of xy^3 and x^3y may be rendered equal.

The solution at first approximation is:

$$x = \frac{\sqrt{\alpha_1}}{n}\cos(nt + 2n\beta_1),$$

$$y = \frac{\sqrt{\alpha_2}}{n}\cos(nt + 2n\beta_2);$$

where $n = \sqrt{2c_1} = \sqrt{2c_2}$.

§ 21. Let us now pass to the simplification of the equations of motion. Corresponding to what was said in § 15 for the case $S=4$ we may here replace in the terms of order h^3 of the equations of motion:

$$\begin{array}{lll} \dot{x}^2 & \text{by} & \alpha_1 - n^2x^2, \\ \dot{y}^2 & \text{,,} & \alpha_2 - n^2y^2, \\ \ddot{x} & \text{,,} & -n^2x, \\ \ddot{y} & \text{,,} & -n^2y. \end{array}$$

The equations become:

$$\left.\ddot{x} + n^2x + 4e_1x^3 + 3e_2x^2y + 2e_3xy^2 + e_4y^3 + \frac{n^4}{g^2}(\alpha_1+\alpha_2)x - \frac{2n^6}{g^2}(x^2+y^2)x = 0.\right|$$

$$\left.\ddot{y} + n^2y + e_2x^3 + 2e_3x^2y + 3e_4xy^2 + 4e_5y^3 + \frac{n^4}{g^2}(\alpha_1+\alpha_2)y - \frac{2n^6}{g^2}(x^2+y^2)y = 0.\right|$$

Here we may omit no terms, for all the terms of order h^3 are disturbing. The equations may be written as follows:

$$\left.\ddot{x} + n^2x - \frac{\partial R}{\partial x} = 0,\right|$$

$$\left.\ddot{y} + n^2y - \frac{\partial R}{\partial y} = 0;\right|$$

where we must take

$$-R \equiv e_1 x^4 + e_2 x^3 y + e_3 x^2 y^2 + e_4 x y^3 + e_5 y^4 + \\ + \frac{n^4}{2g^2}(\alpha_1 + \alpha_2)(x^2 + y^2) - \frac{n^3}{2g^2}(x^2 + y^2)^2.$$

§ 22. If we substitute in the function R for x and y the expressions assumed at first approximation and if we retain only those terms not containing t explicitly, we arrive at

$$-R \equiv \frac{1}{2} a\alpha_1^2 + b\alpha_1\alpha_2 + \frac{1}{2} c\alpha_2^2 + f\alpha_1\alpha_2 \sin^2\varphi + f_1(\alpha_1 + \alpha_2)\sqrt{\alpha_1\alpha_2}\cos\varphi,$$

where

$$a = \frac{3e_1}{4n^4} + \frac{n^2}{8g^2}$$

$$b = \frac{3e_3}{8n^4} + \frac{n^2}{8g^2},$$

$$c = \frac{3e_5}{4n^4} + \frac{n^2}{8g^2},$$

$$f = -\frac{e_3}{4n^4} + \frac{n^2}{4g^2},$$

$$f_1 = \frac{3e_2}{8n^4},$$

$$\varphi = 2n(\beta_1 - \beta_2).$$

The system of differential equations indicating the time-variability of the α's and β's becomes:

$$\left.\begin{aligned}
\frac{d\alpha_1}{dt} &= -4nf\alpha_1\alpha_2 \sin\varphi\cos\varphi + 2nf_1(\alpha_1 + \alpha_2)\sqrt{\alpha_1\alpha_2}\,.\sin\varphi \\
\frac{d\alpha_2}{dt} &= +4nf\alpha_1\alpha_2 \sin\varphi\cos\varphi - 2nf_1(\alpha_1 + \alpha_2)\sqrt{\alpha_1\alpha_2}\,.\sin\varphi, \\
\frac{d\beta_1}{dt} &= a\alpha_1 + b\alpha_2 + f\alpha_2\sin^2\varphi + \frac{1}{2}f_1\left(3\sqrt{\alpha_1\alpha_2} + \alpha_2\sqrt{\frac{\alpha_2}{\alpha_1}}\right)\cos\varphi, \\
\frac{d\beta_2}{dt} &= b\alpha_1 + c\alpha_2 + f\alpha_1\sin^2\varphi + \frac{1}{2}f_1\left(\alpha_1\sqrt{\frac{\alpha_1}{\alpha_2}} + 3\sqrt{\alpha_1\alpha_2}\right)\cos\varphi.
\end{aligned}\right\} \quad (21)$$

It appears at once from the system that:

$$\frac{d\alpha_1}{dt} + \frac{d\alpha_2}{dt} = 0,$$

so

$$\alpha_1 + \alpha_2 = \text{constant}.$$

Another integral is according to § 4:

$$\frac{1}{2} a\alpha_1^2 + b\alpha_1\alpha_2 + \frac{1}{2} c\alpha_2^2 + f\alpha_1\alpha_2 \sin^2\varphi + f_1(\alpha_1 + \alpha_2)\sqrt{\alpha_1\alpha_2}\cos\varphi = const.$$

§ 25. The results become very intricate for the general case. This is evidently a consequence of the circumstance, that in the function R appear $\cos\varphi$ and $\sin^2\varphi$ or in other words $\cos\varphi$ and $\cos 2\varphi$. The problem is considerably simplified if we suppose $f_1 = 0$, thus $e_2 = 0$, which means, that we suppose the planes XZ and YZ to be planes of symmetry for the surface.

Let us again introduce ζ, so that

$$\alpha_1 = R_0^2 h^2 \zeta, \qquad \alpha_2 = R_0^2 h^2 (1 - \zeta),$$

then the last integral may be written in the form:

$$\sqrt{\zeta(1-\zeta)}\cos\varphi = p\zeta^2 + q\zeta + r,$$

so that we can perform again all integrations in finite form, and x and y may then be found as functions of the time.

Osculating curves.

§ 24. We return to the general case and shall proceed to investigate what becomes of the osculating curves. They are ellipses whose equations are found by eliminating t between

$$x = \frac{\sqrt{\alpha_1}}{n}\cos(nt + 2n\beta_1)$$

and

$$y = \frac{\sqrt{\alpha_2}}{n}\cos(nt + 2n\beta_2).$$

By changing the origin of time we see that for a definite osculating curve we can also find the equation by elimination of t between

$$x = \frac{\sqrt{\alpha_1}}{n}\cos nt$$

and

$$y = \frac{\sqrt{\alpha_2}}{n}\cos(nt - \varphi),$$

so φ represents the difference in phase.

When φ has an arbitrary value, the ellipse has an arbitrary shape and position.

If $\varphi = 0$ or π a straight line is described passing through O.

If $\varphi = \frac{\pi}{2}$ the axes of the ellipse lie along the axes of coordinates.

The ellipses are described in rectangles having their sides parallel to the axes and whose vertices, as is evident from

$$a_1 + a_2 = constant,$$

lie on the circumference of a circle.

To investigate the change in shape and position we may write down the well-known relations which may serve for the calculation of the axes of the ellipse and the angle of inclination of the long axis with the X-axis. If Ah and Bh are half the larger and half the smaller axis and if θ is the angle in view, then these relations become:

$$\frac{1}{A^2h^2} + \frac{1}{B^2h^2} = \frac{n^2R_0^2h^2}{a_1a_2\sin^2\varphi}, \quad \ldots\ldots\ldots (1)$$

$$\frac{1}{A^2B^2h^4} = \frac{n^4}{a_1a_2\sin^2\varphi}, \quad \ldots\ldots\ldots (2)$$

$$tg\,2\theta = \frac{2\sqrt{a_1a_2}}{a_2 - a_1}\cdot\cos\varphi. \quad \ldots\ldots\ldots (3)$$

From (1) and (2) we now deduce at once: The sum of the squares of the axes of the ellipse is constant.

§ 25. From what we have just found we can easily prove that in case the surface is a surface of revolution the osculating ellipse has an invariable shape.

Then namely we find:

$$-R = \tfrac{1}{2}a(a_1+a_2)^2 + f\,a_1a_2\sin^2\varphi,$$

where:

$$a = \frac{3e_1}{4n^4} + \frac{n^2}{8g^2},$$

$$f = -\frac{e_1}{2n^4} + \frac{n^2}{4g^2}.$$

As

$$\tfrac{1}{2}a(a_1+a_2)^2 + f\,a_1a_2\sin^2\varphi = constant,$$

and also

$$a_1 + a_2 = constant,$$

we find

$$a_1a_2\sin^2\varphi = constant.$$

From (2) it then follows that

$$ABh^2 = constant,$$

from which in connection with the close of § 24 we may conclude that

$$Ah = const. \quad , \quad Bh = const.,$$

and so our proposition is proved.

If in further consideration of the case of the surface of revolution we wish to see in what way θ varies, we have to write down the differential equations giving the variability of the α's and β's. They now become:

$$\left.\begin{aligned}
\frac{d\alpha_1}{dt} &= -4\,nf\,\alpha_1\,\alpha_2\,sin\,\varphi\,cos\,\varphi\ , \\
\frac{d\alpha_2}{dt} &= \quad 4\,nf\,\alpha_1\,\alpha_2\,sin\,\varphi\,cos\,\varphi\ , \\
\frac{d\beta_1}{dt} &= a\,(\alpha_1+\alpha_2) + f\,\alpha_2\,sin^2\,\varphi\ ; \\
\frac{d\beta_2}{dt} &= a\,(\alpha_1+\alpha_2) + f\,\alpha_1\,sin^2\,\varphi\ .
\end{aligned}\right\}$$

We see that in $\frac{d\beta_1}{dt}$ and $\frac{d\beta_2}{dt}$ an equal constant term $a\,(\alpha_1+\alpha_2) = a\,R_0^{\,2}\,h^2$ appears. This means that the frequency n is modified by an amount of $2\pi a\,R_0^{\,2}\,h^2$.

When we now differentiate according to t the relation

$$tg\,2\theta = \frac{2\sqrt{\alpha_1\alpha_2}}{\alpha_2-\alpha_1}\,cos\,\varphi$$

we may arrive after some reduction at:

$$\frac{d\theta}{dt} = -\,2\,fn^2\,ABh^2\ ,$$

from which it is evident, that the ellipse revolves with a constant angular velocity.

These results agree quantitatively with those found by Prof. KORTEWEG.

§ 26. The change in shape and position of the osculating curve does not seem to become simple for the general case $n_2 = n_1$.

Let us therefore restrict ourselves to the case $e_2 = 0$; then the XZ-plane and the YZ-plane are planes of symmetry for the surface.

The first equation of (21) now becomes

$$\frac{d\alpha_1}{dt} = -\,4\,nf\,\alpha_1\,\alpha_2\,sin\,\varphi\,cos\,\varphi.$$

Or by introduction of ζ:

$$\frac{d\zeta}{dt} = -\,4\,nf\,R_0^{\,2}\,h^2\,\zeta\,(1-\zeta)\,sin\,\varphi\,.\,cos\,\varphi.$$

The relation between ζ and φ becomes:

$$\cos^2\varphi = \frac{p\zeta^2 + q\zeta + r}{\zeta(1-\zeta)} \quad \ldots\ldots\ldots\ldots \quad (22)$$

Here ζ again varies periodically between a greater and a smaller value. Now however $\frac{d\zeta}{dt}$ may become equal to 0 for $\sin\varphi = 0$ and for $\cos\varphi = 0$. Thus barring special cases there are 3 general cases:

1st. For the extreme values of ζ $\cos\varphi = 0$. Then in the extreme rectangles ellipses are described with the axes along the X-axis and Y-axis (fig. 13).

2nd. For the extreme values of ζ $\sin\varphi = 0$. In the extreme rectangles straight lines are described (fig. 14).

3rd. For *one* of the extreme values of ζ $\sin\varphi = 0$, for the other $\cos\varphi = 0$. (fig. 15).

Special cases.

§ 27. These we have again for the extreme values of the modulus $\varkappa$ ($\varkappa$ has the same form as in § 16) of the elliptic functions; which occurs when 2 roots of the equation:

$$f(\zeta) \equiv (p\zeta^2 + q\zeta + r)\{\zeta(1-\zeta) - (p\zeta^2 + q\zeta + r)\} = 0$$

have coincided.

The special case corresponding to B of § 9 and the second of § 18 occurs here in two ways. We refer to the cases in which the same straight line is continually described (continually $\sin\varphi = 0$; when the surface is surface of revolution, this form of motion is possible in every meridian) and that continually the same ellipse is described ($\cos\varphi = 0$; this becomes for the surface of revolution the uniform motion in a parallel circle).

The special case corresponding to A of § 9 and to the first of § 18 exists here too. The form of motion approaches asymptotically the motion in a definite ellipse.

Envelope of the osculating curves.

§ 28. Two cases may be indicated, in which the envelope assumes a simple shape.

1. For $p = -1$, $q = 1$ in (22) (the case of a surface of revolution), the envelope has degenerated into two concentric circles.

2. For $p = 0$ and $q = 0$ in (22) the envelope has degenerated into two pairs of parallel lines, enclosing a rectangle.

Arbitrary mechanism with 2 degrees of freedom for which $S = 2$.

§ 29. The equations of LAGRANGE get quite the same form here

as for $S=4$. In the terms of order h^3 we may in the same way substitute other terms for the terms $\ddot{q}_1$, $\ddot{q}_2$, $\dot{q}_1^2$, and $\dot{q}_2^2$.

Then we have to reduce the terms $-\frac{\partial P_1}{\partial q_2}\dot{q}_1\dot{q}_2$ and $-\frac{\partial P_2}{\partial q_1}\dot{q}_1\dot{q}_2$.

To this end we introduce q'_1 and q'_2 in such a way, that

$$q'_1 = q_1 + \frac{1}{4}a_2 q_1^2 q_2 + \frac{1}{2}a_3 q_1 q_2^2 .$$

$$q'_2 = q_2 + \frac{1}{4}c_1 q_1 q_2^2 + \frac{1}{2}c_2 q_1^2 q_2 .$$

After these reductions it is evident that the terms of order h^3 in the first equation assume the form:

$$\left\{-a_1\frac{\alpha_1}{n^2} + (a_3 - b_2 + c_1)\frac{\alpha_2}{n^2}\right\} q_1 + \left(\frac{1}{2}c_2 - 2b_3\right)\frac{\alpha_2}{n^2}q_2 +$$

$$+ 2a_1 q_1^3 + \left(\frac{1}{2}a_2 + b_1\right)q_1^2 q_2 - (a_3 - 2b_2 + c_1)q_1 q_2^2 - \left(\frac{1}{2}c_2 - 3b_3\right)q_2^3 .$$

We now substitute $\frac{4}{3}q_2^3$ for $B^2h^2q_2$. This is allowed, because substituting $q_2 = Bh\cos(nt+\lambda)$ in $\frac{4}{3}q_2^3$, we obtain besides a term $B^2h^2q_2$ terms which are non-disturbing.

We wish to investigate whether the disturbing terms in the two equations are again derivatives of the same function. For this we need not consider the terms with q_1 and q_1^3, in the first equation and those with q_2 and q_2^3 in the second. The remaining terms become in the first equation:

$$\left(\frac{1}{2}a_2 + b_1\right)q_1^2 q_2 - (a_3 \quad 2b_2 + c_1)q_1 q_2^2 + \frac{1}{3}\left(b_3 + \frac{1}{2}c_2\right)q_2^3 .$$

In the second:

$$\frac{1}{3}\left(\frac{1}{2}a_2 + b_1\right)q_1^3 - (a_2 - 2b_2 + c_1)q_1^2 q_2 + \left(b_3 + \frac{1}{2}c_2\right)q_1 q_2^2$$

So finally we find that the disturbing terms are derivatives of the same function R; so the equations become:

$$\ddot{q}_1 + n^2 q_1 - \frac{\partial R}{\partial q_1} = 0 ,$$

$$\ddot{q}_2 + n^2 q_2 - \frac{\partial R}{\partial q_2} = 0 ,$$

where $$R = Ph^2 q_1^2 + Qh^2 q_2^2 + U_4 ,$$

when P and Q are homogeneous quadratic functions of $\sqrt{\alpha_1}$ and $\sqrt{\alpha_2}$ and when U_4 is a homogeneous function of order four of q_1 and q_2. The results found for the simple mechanism hold therefore for an arbitrary mechanism with two degrees of freedom.

Mathematics. — *"The cubic involution of the first rank in the plane."* By Dr. W. VAN DER WOUDE. (Communicated by Prof. P. H. SCHOUTE.)

1. If V is a plane it is in different ways possible to arrange the points of V in groups of three in such a way, that an arbitrary point forms a part of only one group. If P_1 is a point of V there must exist between the coordinates of P_1 and those of the other points of the group, to which P_1 belongs, some relations by which those other points are entirely determined. It is however possible that P_1 can be chosen is such a way that one of these relations is identically satisfied; in that case P_1 forms part of an infinite number of groups.

We now start from the following definition:

The points of a plane V form a cubic involution of the first rank, when they are conjugate to each other in groups of three in such a way that (with the exception of some definite points) each point forms a part of only one group.

A triangle of which the vertices belong to a selfsame group we call an *involution triangle*; each point which is a vertex of more than *one*, therefore of an infinite number of involution triangles, we call a *singular point* of the involution; each point coinciding with one of its conjugate points is called a *double point*. If one of the sides of an involution triangle rotates around a fixed point, then the third vertex of this triangle will describe a right line or a curve; *we shall restrict ourselves in this investigation to the case, that one vertex of an involution triangle describes a right line, when the opposite side rotates around a fixed point.*

2. When the points of a plane V form a cubic involution of the first rank which satisfies the just mentioned condition and which we shall furtheron indicate by (i_3), we can conjugate projectively to each point of V the connecting line of its conjugate points. Each vertex of an involution triangle and its opposite side are pole and polar line with respect to a same conic, which in future we shall always call γ_2; each involution triangle is a polar triangle of γ_2. It is clear that reversely not every polar triangle of γ_2 is an involution triangle of (i_3); for each point of V is a vertex of an infinite number of polar triangles of γ_2, but of only *one* involution triangle. If however S is a singular point of the involution, then S must be a vertex of an infinite number of involution triangles, thus each polar triangle of γ_2 having S as vertex is at the same time an

involution triangle. If we assume a point G of the conic γ_2 as a vertex of an involution triangle, then one of the other vertices must coincide with G, so G is a double point of the involution; γ_2, the locus of these double points, is the *double curve* of the involution.

Each line l whose pole with respect to γ_2 is no singular point of the involution is a side of only one involution triangle, namely of that triangle having the pole of l as vertex. On the other hand each line whose pole is a singular point is a side of an infinite number of involution triangles all having that point as vertex. From this ensues that also the lines of V form a cubic involution (i'_3) of the first rank; the polar lines of the singular points of (i_3) are the singular lines of (i'_3), the tangents of γ_2 are its double lines and γ_2 is its double curve. Both involutions are with respect to γ_2 polarly related.

The involution triangles of γ_2 are all polar triangles of a selfsame conic γ_2, which is at the same time the double curve of (i_3). The lines of V form an involution (i'_3) which is with respect to γ_2 the polar figure of (i_3). Each polar triangle of γ_2 having a singular point of the involution as vertex is at the same time an involution triangle.

3. We make a point describe a line a_1 and we ask after the locus of its conjugate points. If we draw through A_1, the pole of a_1 with respect to γ_2, an arbitrary line p_1, then P_1, the pole of p_1, lies on a_1, whilst the two points conjugate to P_1 lie on p_1; these two points lie also on the locus under discussion. Moreover A_1 itself is conjugated to two points of a_1, so that A_1 is a double point of this curve and each line through A_1 cuts this curve in a double point and two points more. Hence we find:

If one of the vertices of an involution triangle describes a line a_1, then the two others describe a curve α^4 of order four with a node in A_1, the pole of a_1 with respect to γ_2. As a_1 cuts all singular lines, all singular points lie on α^4.

A few properties of this curve α^4 may still be given here:

1. Let A_2 and A_3 be the points conjugated to A_1, then the polar line of A_3 with respect to γ_2 — that is the line A_1A_3 — must cut α^4 in A_1 and in the points forming with A_3 an involution triangle. These two points are A_1 and A_2. So will α^4 be touched in A_1 by the lines A_1A_2 and A_1A_3; A_2 and A_3 are points of intersection of a_1 and α^4.

2. Besides in A_2 and A_3 the curve α^4 will be intersected in two points more by a_1; these points are at the same time the points of intersection of a_1 and γ_2.

3. Besides in these last points α^4 will still be cut by γ_2 in 6 points more, the tangents in these 6 points to α^4 must pass through A_1. From this ensues that α^4 is of the tenth class, by which the PLÜCKER numbers of α^4 are entirely determined ($n=4, m=10, d=1$). This holds, for it is easy to investigate that α^4 cannot possess a double point differing from A_1.

4. If a vertex of an involution triangle describes a line, on which lies a singular point, the curve described by the two other vertices degenerates into the polar line of that singular point and a curve which must be of order three. If a vertex of an involution triangle describes a singular line s, then one of the other two vertices will be a fixed point, namely the pole of s and the other point will describe s itself and as many other lines as there are singular points on s. As both points together must describe a curve of order four, three singular points will lie on s. In like manner each singular point is point of intersection of three singular lines.

If now again a_1 is an arbitrary line and if α^4 has the same signification as above, then the curve α^4 will cut a line b_1 four times; from this ensues that four times a point of a_1 and a point of b_1 are vertices of a selfsame involution triangle. These vertices we call P_1, Q_1, R_1, S_1 and P_2, Q_2, R_2, S_2, whilst the third vertices of these triangles may be represented by P_3, Q_3, R_3, S_3; farthermore T_1 is the point of intersection of a_1 and b_1 and T_2 and T_3 are the two points forming with T_1 an involution triangle.

If now a point describes the line b_1, then its conjugate points describe a curve β^4 of order four; α^4 and β^4 have 16 points of intersection. These are:

1. the two points T_2 and T_3;
2. the four points P_3, Q_3, R_3, S_3;
3. ten points more having the property that to each of them two, so an infinite number of pairs of points, are conjugated and which are thus the singular points. Therefore:

The involution (i_3) *has* 10 *singular points; their polar lines are the* 10 *singular lines of* (i'_3).

These singular elements have such a position that on each of these lines three of these points lie and that in each of the points three of the lines intersect each other; so they form a configuration $(10_3, 10_3)$.

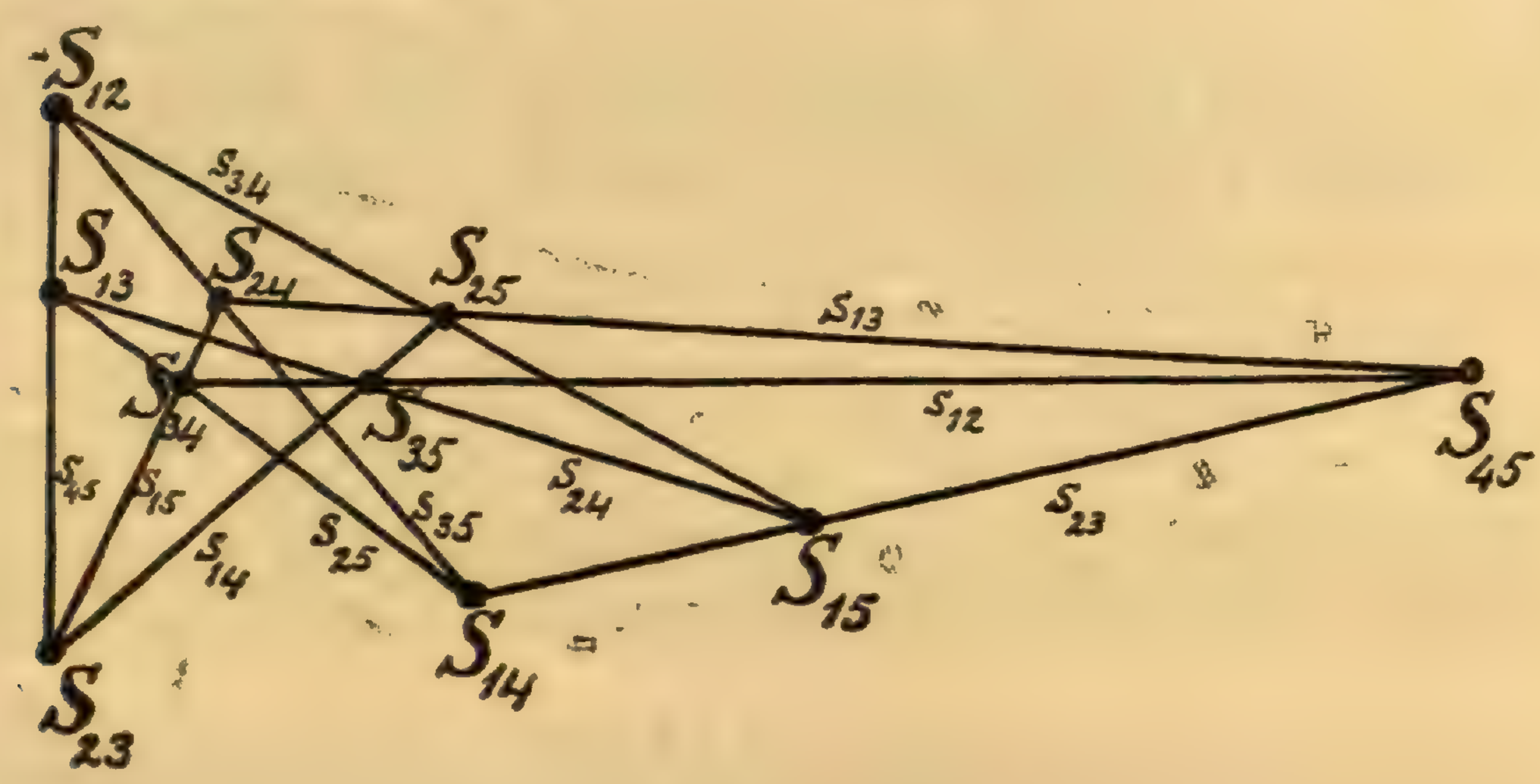
If s_{12} is a singular line and S_{12} its pole with respect to γ_2, then there are besides S_{12} still 6 singular points not lying on s_{12}. If S_{13} is one of these points and s_{13} the polar line of S_{13}, then the point of intersection of s_{12} and s_{13} is at the same time the pole of $S_{12}S_{13}$.

This point forms an involution triangle with S_{12} and with another point of s_{12} and an *other* one with S_{13} and with a point of s_{13} (an "other one", as S_{12} and S_{13} which do not lie on each other's polar line cannot be vertices of a selfsame involution triangle); so the point of intersection of s_{12} and s_{13} is also a singular point and $S_{12}S_{13}$ a singular line.

Each line connecting two singular points not lying on each other's polar line is a singular line; each point which is the point of intersection of two singular lines not passing through each other's pole is a singular point.

On s_{12}, the polar line of S_{12}, lie 3 singular points; the remaining 6 are connected with S_{12} by 3 singular lines. So each line connecting S_{12} with one of the singular points on s_{12} is not a singular line, as only 3 of these lines pass through S_{12}.

We can indicate the position of the singular points by the following diagram, where the indices have been chosen in such a way that always the points S_{ik}, S_{kl} and S_{il} lie on a selfsame line, that the lines s_{ik}, s_{kl} and s_{il} intersect each other in a selfsame point, and that the point S_{ik} and the line s_{ik} are each other's pole and polar line with respect to γ_2.



5. We make a point describe a conic α_2 and an other point a line b_1 the two points which are conjugated to the former describe a curve α^n, those which are conjugated to the latter a curve β^4. As β^4 and α_2 intersect each other in 8 points, b_1 and α^n must have 8 points in common, so α^n is a curve of order eight; we shall call it in future α^8. As α_2 intersects all singular lines twice, α^8 will have in each of the 10 singular points a node.

If α_2 is described around an involution triangle, then α^8 has also double points in the vertices of this triangle. As all involution

.riangles are at the same time polar triangles of a selfsame conic γ_2, we can describe a conic around each pair of involution triangles; if a conic β_2 is described around two of these triangles, then the curve β^8 conjugate to it will have 6 nodes in its circumference. Also the remaining points of intersection of β_2 and β^8 are easily indicated; they are the four points of intersection of β_2 and γ_2.

We know moreover that a conic described around an involution triangle and through two of the vertices of an other involution triangle must also contain the third vertex of the latter.

6. It is also clear, that we can easily construct conics described around three involution triangles; to that end we make a conic, pass through the vertices of an arbitrary involution triangle and through two singular points not lying on each other's polar line; for this we choose S_{12} and S_{13}. As α_2 is described around a polar triangle of γ_2, it is described around an infinite number of these triangles; further *each* polar triangle of γ_2 having one of the singular points as vertex is at the same time an involution triangle, so that α_2 is described around three involution triangles.

Now the curve α^8 will have in the circumference of α_2 nine nodes; so it must degenerate and α_2 must be one of the parts into which it breaks up. If P_1 is an arbitrary point of α_2 then always one of the two points P_2 and P_3 forming with P_1 an involution triangle will also lie on α_2, so also the third vertex lies on α_2 (5). If now we let P_1 describe the conic α_2, then P_2 and P_3 will describe the same curve; every time however that P_1 coincides with one of the singular points on α_2, P_2 and P_3 will be bound to no other condition, than that they must lie on the polar line of that point and must form with P_1 a polar triangle of γ_2. So the parts into which α^8 degenerates are:

1. the conic α_2 to be counted double;

2. as many lines as there are singular points lying on α_2.

From this ensues that besides S_{12} and S_{13} 2 more singular points lie on α_2.

This last we can prove still in another way; we construct a second conic β_2, described around an involution triangle $Q_1\ Q_2\ Q_3$ and through S_{12} and S_{13}; it will cut α_2 in two points more, which being both the vertices of two, i.e. of an infinite number of involution triangles, are therefore singular points. If we construct another conic δ_2 described around a triangle of involution $R_1\ R_2\ R_3$ and through S_{12} and S_{13}, then this must still cut α_2 in two singular points; these

must be the same as the points of intersection of α_2 and β_2, because on α_2 no more than four singular points can lie.

So all conics passing through S_{12} and S_{13} and farthermore described around *one*, hence around an infinite number of involution triangles will form a pencil; the two other base points of this pencil are also singular points. We determine these first: if we choose as β_2 the pair of lines S_{24} and S_{35} and as δ_2 the pair S_{34} and S_{25}, it is evident that S_{14} and S_{15} are the discussed base points. Therefore: *If the* 10 *singular points, hence also the double curve* γ_2, *of the involution are known, we can generate the involution triangles in this way:*

We can construct five different pencils of conics of which each conic is described around an infinite number of polartriangles of γ_2, *which are then at the same time the involution triangles in view; the base points of these pencils consist of the sets of points* (S_{12}, S_{13}, S_{14}, S_{15}), (S_{12}, S_{23}, S_{24}, S_{25}), (S_{13}, S_{23}, S_{34}, S_{35}), (S_{14}, S_{24}, S_{34}, S_{45}) and (S_{15}, S_{25}, S_{35}, S_{45}).

These pencils we shall call in future respectively (B_1), (B_2), (B_3), (B_4) and (B_5).

If α_1 and α_2 are two conics, the first taken arbitrarily out of (B_1), the second arbitrarily out of (B_2), these two will have four points of intersection, viz. S_{12} and the vertices of an involution triangle. Now it can happen in two different ways that 2 of these points of intersection coincide: 1. S_{12} can be at the same time a vertex of the involution triangle 2. one of these vertices can lie on the double curve γ_2. In each of these two cases α_1 and α_2 will have only three different points in common, but they will touch each other moreover in one of these points.

7. Out of these 5 pencils we choose one — e.g. (B_1) — arbitrarily; an arbitrary conic δ_2 out of (B_1) is described around an infinite number of involution triangles whose vertices form in its circumference an involution of order three. The latter has four double points in the points of intersection of δ_2 with γ_2, the double curve of the involution (i_3). Inversely the conics of the pencil (B_1) determine an involution of order four on γ_2; the latter has 6 double points in the points in which γ_2 is touched by a conic out of (B_1). In each of these points three points have thus coincided, forming together a group of (i_3).

The involution (i_3) has 6 triple points; in each of the points γ_2 is touched by a conic out of each of the pencils (B_1), (B_2), (B_3), (B_4), and (B_5).

8. A point whose conjugate points coincide we call a *branch point*, the locus of these points the *branch curve*. If we let a point G describe the conic γ_2, then the curve of order eight, generated by the points conjugate to G, must degenerate into 2 parts, of which one is γ_2 itself and the other the branch curve. From this ensues that the latter is of order six and possesses nodes in the 10 singular points; so it is rational as it should be, as it corresponds point for point to a conic.

Also in an other way we can easily deduce the order of the branch curve; if a point describes a line a_1, then the conjugate points describe a curve α^4 having with γ_2 eight points of intersection, of which two coincide with the points of intersection of a_1 and γ_2, whilst the others point to 6 points of intersection of a_1 with the branch curve.

If G_{12} is a point of the double curve γ_2 and g the tangent in that point to γ_2, then g will intersect the branch curve in 6 points of which one G_3 forms with the double point G_{12} a group of conjugate points; so in the triple points of the involution γ_2 and the branch curve will have to touch each other.

The branch curve is a rational curve of order six, having double points in the singular points and touching the double curve in the triple points of the involution.

Observation. A rational curve of order six has 10 double points; of which however only 8 can be taken arbitrarily [1]); *from the preceding follows however that* 10 *points determining a Cf* $(10_3, 10_3)$ *can always be double points of a rational curve of order six.*

In an other form C. F. GEISER (see his paper quoted in the following number) makes the same observation.

9. We shall now apply the preceding to some problems out of Threedimensional Geometry. To that end we regard the pencil (B) of twisted cubics which can be brought through 5 fixed points P_1, P_2, P_3, P_4, and P_5. These determine on an arbitrary plane V a cubic involution of rank one; the lines $P_i\ P_j$ cut V in the singular points S_{ij}, the planes $P_k\ P_l\ P_m$ cut V in the singular lines s_{ij} of the involution. Through an arbitrary point of V passes only one curve out of this pencil, through a singular point S_{ij} however pass an infinite number of curves, which have all degenerated into the fixed line $P_i P_j$ and a variable conic; these conics form a pencil with P_k, P, P_m and the point of intersection of $P_i P_j$ with the plane $P_k P_l P_m$ as base points. Each double point of the involution in V is now a point, in which a twisted curve out of the pencil (B)

[1]) SALMON-FIEDLER: Höhere ebene Kurven, Zweite Auflage, p. 42.

touches the plane V; the third point of intersection of this curve with V is a point of the branch curve forming with the point of contact a group of mutually conjugate points of the involution. A triple point of the involution is a point, in which a twisted curve out of (B) is osculated by V. From this ensues:

1. *All twisted cubics passing through* 5 *given points and touching a given plane* V *form a surface* F^{10} *of order ten, which touches* V *in a conic and cuts* V *moreover according to a rational curve of order six.*

2. *There are* 6 *twisted cubics passing through five given points and having a given plane as osculating plane.*

As a special case of this last theorem we have still: *through five given points pass six twisted parabolae.*

Through the pencil (B) of twisted cubics with P_1, P_2, P_3, P_4 and P_5 as base points a plane V is cut according to a cubic involution of the first rank. If α is a curve out of this pencil cutting V in A_1, A_2 and A_3, then α is projected out of A_1 by a cone cutting V according to the lines $A_1 A_2$ and $A_1 A_3$. If however a curve γ out of (B) touches a plane V in a point G_{12} and if moreover it cuts V in a point G_3, then γ is projected out of G_{12} by a cone cutting V according to $G_{12} G_3$ and the tangent in G_{12} to γ; γ is projected out of G_3 by a cone touching V according to $G_3 G_{12}$. We have seen that G_{12} must lie on the double curve and G_3 on the branch curve of the involution, whilst $G_3 G_{12}$ touches the former; if therefore a quadratic cone is to pass through the base points of the pencil (B) and to touch V moreover, then its vertex must lie on the branch curve and the tangent with V must touch the double curve.

The number of quadratic cones passing through five given points and touching a given plane is singly infinite; the tangents envelope a conic. The vertices of the cones form a rational curve of order six.[1])

The tangential planes of all these cones whose number is ∞^2 envelope a surface of which we wish to determine the class and which for the present we will call Φ_n. If K_2 is one of these cones and G_3 its vertex, then through a line l drawn in V through G_3 one more tangential plane to K_2 will pass; as l has with the branch curve 6 points of intersection, it lies still in 6 tangential planes of Φ_n except in V. Farthermore V is a trope of Φ_n (that is a tangential plane touching (γ_2) in the points of a conic) to be counted double; the surface Φ_n is therefore of class eight.

The tangential planes of these cones envelope a surface of class eight[1])

[1]) C. F. Geiser: "Uber Systeme von Kegeln zweiten Grades".

We finally put the question how many twisted circles can be brought through five points where we understand by a twisted circle a twisted cubic cutting the isotropic circle in two points. All twisted cubics through these five points describe on the plane at infinity an involution; if now a point describes the isotropic circle, its conjugate points will describe a curve of order eight having with this circle sixteen points in common; four of these points are at the same time double points of the involution, whilst the other lie two by two on a same twisted circle.

So through five given points pass ten twisted circles, of which four touch the plane at infinity.

Mathematics. — "*On the surfaces the asymptotic lines of which can be determined by quadratures*". By J. BRUIN. (Communicated by Prof. HK. DE VRIES).

In a paper entitled as above A. BUHL (*Nouv. Ann. de Math.*, 4e série, vol. 8, page 433, vol. 9, page 337, *Rev. sem.* XVII 2, page 62, XVIII 1, page 58) discusses the surfaces given by the parameter representation

$$x = r \cos \theta,$$
$$y = r \sin \theta,$$
$$\Phi(z) = a\,\theta + F(r),$$

in which x, y, z refer to a rectangular system of coordinates, so that z, θ, and r are the so-called cylindric coordinates; these are the only ones which are used in the course of the investigation.

BUHL now gives the differential equation of the asymptotic lines of $\Phi(z) = a\,\theta + F(r)$ with θ and r as independent variables as well as with z and θ. It is then evident that this equation embraces many special cases, where the determination of the asymptotic lines comes to quadratures.

We can put the question more in general: which are the surfaces of one of the forms $z = \varphi(r, \theta)$, or $\theta = f(r, z)$, or $r = f(z, \theta)$, whose asymptotic lines can be determined by quadratures?

Starting from the differential equation of the asymptotic lines

$$D\,du^2 + 2\,D'\,du\,dv + D''\,dv^2 = 0$$

(BIANCHI-LUKAT, "Vorlesungen über Differentialgeometrie", page 109), where D, D' and D'' have the values, to be found on page 87 of the quoted work, we find for the differential equation in r and θ of the asymptotic lines of $z = \varphi(r, \theta)$:

$$r\frac{\partial^2 z}{\partial r^2}\,dr^2 + 2\left(r\frac{\partial^2 z}{\partial r\,\partial\theta} - \frac{\partial z}{\partial\theta}\right)dr\,d\theta + \left(r\frac{\partial^2 z}{\partial\theta^2} + r^2\frac{\partial z}{\partial r}\right)d\theta^2 = 0 \quad . \quad (1)$$

This equation gives rise to quadratures in the following cases:

a. $z = a\,\theta + f(r)$, BUHL, l. c., vol. 8, p. 439, comp. also TISSERAND, "Rec. compl. d'exercices", p. 426.

b. $l(z) = a\,\theta + f(r)$, BUHL, l. c. p. 440.

c. $z = A\,r\,sin\,(\theta + \alpha) + a\theta + F(r)$,

d. $z = r\,f_1(\theta) + f_2(\theta)$,

e. $z = r^k\,sin\,(\theta\sqrt{k} + c)$,

f. $z = r^{-k}\,e^{\theta\sqrt{k}+c}$,

g. $z = -\theta^2 + 2\,l(r)$.

2. The differential equation of the asymptotic lines of $\theta = f(r,z)$ in r and z we find by eliminating θ between (1) and $\theta = f(r, z)$.

We find:

$$\left\{r\frac{\partial^2 f}{\partial r^2} + 2\frac{\partial f}{\partial r} + r^2\left(\frac{\partial f}{dr}\right)^3\right\}dr^2 + 2\left\{r\frac{\partial^2 f}{\partial z\partial r} + r^2\left(\frac{\partial f}{\partial r}\right)^2\frac{\partial f}{\partial z} + \frac{\partial f}{\partial z}\right\}dr\,dz +$$

$$+ r\left\{\frac{\partial^2 f}{\partial z^2} + r\frac{\partial f}{\partial r}\left(\frac{\partial f}{\partial z}\right)^2\right\}dz^2 = 0.$$

This equation gives rise to quadratures in the following cases:

a. $\theta = l(r) + f(z)$,

b. $\theta = arc\,cos\,\frac{k}{r} + f(z)$, Buhl, l. c., vol. 9, p. 343,

besides a few others mentioned above.

3. In an analogous way we find the differential equation of the asymptotic lines in z and θ of $r = f(z, \theta)$.

It runs:

$$\frac{\partial^2 f}{\partial z^2}\,dz^2 + 2\left\{\frac{\partial^2 f}{\partial z\,\partial\theta} - \frac{1}{f}\frac{\partial f}{\partial\theta}\frac{\partial f}{\partial z}\right\}dz\,d\theta + \left\{\frac{\partial^2 f}{\partial\theta^2} - f - \frac{2}{f}\left(\frac{\partial f}{\partial\theta}\right)^2\right\}d\theta^2 = 0.$$

Besides in the above mentioned cases this equation gives rise to quadratures for $r = f_1(z)\,f_2(\theta)$, surfaces of JAMET (Ann. de l'école norm. sup., 1887, Suppl., page 50 etc.; further: PICARD, "Traité d'analyse" I, 2nd ed., page 433).

The classes of surfaces found above are not strictly separated; some even are to be regarded as subclasses of others. They can be ranked according to the most general types to be found among them, whilst others fall under these types, namely as follows:

$z = Ar \sin(\theta + \alpha) + a\theta + F(r)$ $\qquad$ $z = a\theta + F(r)$

$z = rf_1(\theta) + f_2(\theta)$

$r = f_1(z)\, f_2(\theta)$ $\qquad$ $l(r) = \theta + f(z)$

$z = r^k \sin(\theta\sqrt{k} + c)$

$z = r^{-k}\, e^{\theta\sqrt{k}+c}$

$z = -\theta^2 + 2l(r)$

$l(z) = a\theta + f(r)$

$\theta = arc\cos\frac{k}{r} + f(z).$

II. Let us discuss one of the above mentioned classes more closely, viz.

$$z = r f_1(\theta) + f_2(\theta)\ ^1). \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

It is the general equation of the scrolls with the z-axis as directrix. Of these scrolls we can find the striction line in the following way.

BIANCHI (l.c. p. 223) deduces that the curvature $K = \frac{1}{r_1 r_2}$, for which in another place was found $K = \frac{DD'' - D'^2}{EG - F^2}$, is larger in the central point than in all other points of a generatrix. If we make up K for (2) we find

$$K = \frac{-(f_2')^2}{[r^2\{1 + (f_1)^2 + (f_1')^2\} + 2rf_1' f_2' + (f_2')^2]^2}.$$

Along a generatrix θ is constant; there only the denominator of the expression for K changes. If we determine the value of r for which K becomes maximal we find

$$r = -\frac{f_1' f_2'}{1 + (f_1)^2 + (f_1')^2}. \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

So this is the (r,θ) projection of the striction-line.

This equation can be found in an other way, too. We have the property that the tangential plane in the central point of a generatrix is normal to the tangential plane in the point at infinity of that generatrix. We now determine for $f = -z + rf_1(\theta) + f_2(\theta)$ the values of $\frac{\partial f}{\partial x}, \frac{\partial f}{\partial y}, \frac{\partial f}{\partial z}$ in an arbitrary point p and in the point at infinity on the same generatrix. If then p is to be central point the sum of the products $\left(\frac{\partial f}{dx}\right)_p \cdot \left(\frac{\partial f}{\partial x}\right)_\infty$ must be equal to 0. This gives again the equation (3).

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[1]) In future we shall write f_1 and f_2 for $f_1(\theta)$ and $f_2(\theta)$.

Let us now consider in how far we can find the surface (2) for a given striction line.

a. Let $r = \varphi(\theta)$ be the given projection of the striction line. This furnishes, regarding (3), the relation between f_1 and f_2:

$$-\frac{f_1' f_2'}{1 + (f_1)^2 + (f_1')^2} = \varphi$$

$$f_2' = \frac{-\varphi\{1 + (f_1)^2 + (f_1')^2\}}{f_1'}.$$

Thus: The surface $z = r f_1(\theta) + \int \frac{-\varphi\{1 + (f_1)^2 + (f_1')^2\}}{f_1'} d\theta$, where f_1 is an arbitrary function of θ, has as (r, θ)-projection of the striction line the curve $r = \varphi(\theta)$.

b. Let now be given that the striction line must be a plane curve lying in the plane $z = Ax + By + C$ or $z = r(A\cos\theta + B\sin\theta) + C$.

By substituting this value for z in (2) we must get (3). This furnishes between f_1 and f_2 the relation

$$\frac{f_2 - C}{f_1 - A\cos\theta - B\sin\theta} = \frac{f_2' f_1'}{1 + (f_1)^2 + (f_1')^2}$$

or

$$\frac{f_2'}{f_2 - C} = \frac{1' + (f_1)^2 + (f_1')^2}{f_1'(f_1 - A\cos\theta - B\sin\theta)}.$$

So the surface $z = r f_1(\theta) + e^{\int \frac{1 + (f_1)^2 + (f_1')^2}{f_1'(f_1 - A\cos\theta - B\sin\theta)} d\theta} + C$, in which f_1 is an arbitrary function of θ, has a plane striction line lying in $z = Ax + By + C$.

c. The most general problem here is: what is the surface (2) for which $r = \varphi(\theta)$, $z = \psi(\theta)$ is striction line?

To solve this we substitute these values for r and z in (2) and (3) and we obtain then two equations with f_1 and f_2 as unknown quantities. If we eliminate between these two f_2, we retain f_1 as only unknown quantity in the equation:

$$\varphi + q(f_1)^2 + f_1'\psi' - \varphi' f_1 f_1' = 0.$$

If f_1 is solved out of this we can find f_2 out of:

$$\psi = \varphi f_1 + f_2.$$

We can find the solution in explicit form for the special case $\psi = const.$, for which constant we take 0. We then find:

$$\varphi\{1 + (f_1)^2\} - \varphi' f_1 f_1' = 0,$$

from which ensues

$$\pm f_1 = \sqrt{e^{2\int \frac{\varphi}{\varphi'} d\theta} - 1},$$

whilst further

$$f_2 = -\varphi f_1.$$

The result is therefore:

The surface

$$\pm z = (r - \varphi)\sqrt{e^{2\int \frac{\varphi}{\varphi'} d\theta} - 1}$$

has the plane striction line $r = \varphi$, $z = 0$.

The formulae deduced above hold for the surfaces (2). As was noticed the general types mentioned at the conclusion of I are not strictly separated, however, so that there are still amongst them scrolls with a right directrix, to which then the above formulae are applicable.

Examples of this are:

Of the type $z = Ar\sin(\theta + \alpha) + a\theta + F(r)$

the surface $z = Ar\sin(\theta + \alpha) + a\theta + br$.

Of the type $l(z) = a\theta + f(r)$

the surface $l(z) = a\theta + l(r + p)$;

these have the z-axis as directrix.

Of the type $r = f_1(z) f_2(\theta)$ the scrolls

$$x^n(z + 1) = z^n y, \text{ or } r = (tg\,\theta)^{1/n-1} \sec\theta \left(\frac{z^n}{z+1}\right)^{1/n-1}$$

and

$$x^n = (y - cx)z^n \text{ or } r = \sec\theta\,(tg\theta - c)^{\frac{1}{n-1}} z^{\frac{n}{n-1}}$$

have still the y-axis as directrix.

Physics. — "*A new theory of the phenomenon allotropy.*" By Prof. A. Smits. (Communicated by Prof. A. F. Holleman.)

Introduction.

In two short communications inserted in the "Chemisch Weekblad" **7**, 79 and 155 (1910) I have already sketched the main lines of the theory, an extension and experimental confirmation of which follow here.

Before passing on to this I may, however, be allowed to give the gist of this theory in a few words.

In the investigation of the phenomenon tautomerism it has been possible to show by means of the process of solidification that the liquid phases of tautomeric substances are composed of two kinds of molecules.

Besides, however, by deposition of different solid substances the complexity of a liquid phase can also be shown in another way, and investigations in this direction have led to the result that it may be considered as the rule that the liquid phase of a substance is built up of different kinds of molecules (ions included).

BANCROFT [1]) and BAKHUIS ROOZEBOOM [2]) have pointed out that when a substance behaves as a unary substance, this is accounted for by the fact that the setting in of inner equilibrium takes place so rapidly in the homogeneous phase that the inner equilibrium if disturbed, is immediately restored by the appearance or disappearance of a new phase; the melting-point, boiling-point, critical-point etc. of a substance which behaves as a unary one, does not relate then to a *single* kind of molecules, but to an equilibrium between *different* kinds of molecules.

BANCROFT's pupils, viz. CARVETH, SOCH, and CAMERON [3]) have investigated different tautomeric substances; it then appeared that it may be pretty easily shown in some cases that under certain circumstances the existence of two kinds of molecules in the liquid phase may lead to a binary behaviour, for when the liquid cooled *rapidly*, the inner equilibrium could not follow the temperature, and the mass solidified at a temperature which differed from the unary stable melting-point, for then a point was realized of one of the melting-point lines of the pseudo-binary system $A + B$, which for the examined substances always showed a eutectic point.

As is evident we find the unary stable melting-point where the curve for the inner liquid-equilibrium meets one of the melting-point curves of the pseudo-binary system.

Now it is remarkable, as I already wrote, that nobody has observed what surprising results are arrived at when it is assumed, what is undoubtedly true, that not only mixed crystals are always formed in a greater or less degree, but that moreover the *inner* equilibrium, which exists in the liquid phase, continues to exist in the solid phase.

Starting from this supposition we get the relation between *heterogeneous* and *homogeneous* allotropy, indicated in Fig. 1, from which it appears that the phenomenon of enantiotropy means *unmixing* in the solid state, which phenomenon appears when the curves for

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[1]) Journ. Phys. Chem. **2**, 143 (1898).
[2]) Zeitschr. f. phys. Chem. **28**, 289, (1899).
[3]) Journ. Phys. Chem. **2**, 159 (1898).
ibid. **2**, 364 (1898).
ibid. **2**, 409 „

the stable and metastable solid equilibria s_2q and s_1n meet the mixed crystal lines ep and dm of the pseudo-binary system.

In case of monotropy these meetings between the unary and the pseudo-binary system do not take place *under* but *above* the unary melting-point temperature, and this is the reason that in this case the second line for the solid inner equilibria everywhere indicates metastable states.

I started from GIBBS' principle of equilibrium, which states that with constant temperature and pressure a number of substances arranges itself in such a way that the thermodynamic potential is a minimum; and then I showed how sharply the relation between the pseudo-binary and the unary system can be defined also in this way, when we bear in mind that a state of inner equilibrium must always lie in the minimum of a potential line.

Further the case was considered that the three-phase temperature lies between the melting points of the substances A and B. After having discussed the phenomena of enantiotropy and monotropy also for this case, I finally pointed out that when A and B are miscible in all proportions in the solid state, *heterogeneous allotropy* is excluded, and only homogeneous allotropy can occur, *unless* unmixing occurs in the pseudo-binary system at lower temperature.

Discussion of the curves of inner equilibrium.

After this introduction which seemed indispensable to me to make the reader acquainted with the main facts, I will consider fig. 1 a little more closely and discuss the curves of inner equilibrium.

It follows from the course of the curve $S'_3S'_1$ that it is assumed here that over the corresponding range of temperature the equilibrium shifts towards B with increase of temperature, and so that

$$A \rightleftarrows B - a\ cal$$

or in words that thé transformation from the left to the right is endothermic.

With application of the equation:

$$\frac{d\ln K}{dt} = \frac{Q}{RT^2}$$

we know therefore that Q is positive in the assumed case.

Neglecting the external work we can split up Q into two differential heats of mixing, one of which has the negative sign, because it is a case of unmixing, and further into a heat of transformation, so:

$$Q = -(Q_m)_A + Q_r + (Q_m)_B$$

$(Q_m)_A$ = differential heat of mixing of A

$(Q_m)_B$ = „ „ „ „ „ B

Q_r = mol. heat of transformation.

It is of importance to point out here that as $(Q_m)_A$ and $(Q_m)_B$ have a different sign, the *possibility* exists that Q has another sign than Q_r; this might e.g. occur when Q_r was very small, and then we should have the special case that e.g. when Q was negative and Q positive, the equilibrium shifted from B to A with rise of temperature, whereas the transformation of A into B is endothermic in itself; this, however, will only rarely occur.

If we drop this question for the present, it is noteworthy that in the point S'_1 unmixing occurs, another solid phase S'_2 appearing by the side of S'_1. Two cases may be distinguished here.

Generally the newly-formed solid phase S'_2 will possess another form of crystal than S'_1, but it is possible that the two solid phases are isomorphous, for as is known, also isomorphous substances can show partial miscibility; if this latter, the simplest case occurs, the heat of transformation will be the sum of a heat of unmixing, a heat of transformation, and a heat of mixing[1]), another thermal quantity being added to this, viz. that which accompanies the change of crystalline form, when S'_1 and S'_2 are not isomorphous.

If we now follow the inner equilibria *above* the transition-point, it is to be expected that the curves $S'_2 q$ for the solid-, and $l_2 k$ for the liquid inner equilibria will have the same direction as $S'_3 S'_1$, as is also assumed in fig. 1.

Soch, however, has found in his investigation of *benzile-orthocarbonic acid* that the curve of the inner liquid equilibrium meets the melting-point curve of the modification with the highest melting-point viz. B, and runs to the A-side for higher temperatures. Further he found that at 65° A passes into B, and combining these two facts, he arrives at the conclusion that the thermal sign of the transformation

$$A \rightarrow B$$

must have been reversed between the point of transition and the unary melting-point (137°).

When the pseudo-binary T,x-figure for this substance agrees with fig. 1, which is still an open question, we must of course come to the same result also going by this theory, but I will point out here that this conclusion is not yet imperative at this moment, because though it is not probable, the possibility exists that the mixed crystal

[1]) I shall discuss this and the before-mentioned splitting up more fully later on.

curve dm of the pseudo-binary system has the same direction as ep; in this case the three curves of equilibrium $S'_3 S'_1$, $S'_2 S_2$ and $l_2 k$ might still have the same direction, and so the sign of Q need not be reversed.

If we look upon the question of the reversal of the thermal sign from a general point of view, the following may already be remarked. When A and B are *isomers*, as for benzile-orthocarbonic acid, a reversal of the sign of Q seems possible, because Q_r is probably small in this case [1]). If, however, we have to do with the phenomenon polymerism, we may expect with great probability that Q_r will always predominate, and that the curves for the inner solid and liquid equilibria will always run in such a way that the equilibrium shifts towards the side of the less complex substance with rise of temperature.

This leads us at the same time to the question what the T,x-figure will be for the case that the substance B is a *polymer* of A, and that a transition point exists.

Fig. 2 shows that when the pseudo-binary system possesses a eutectic point, the curve for the inner liquid equilibria must meet the melting-point curve of the less complex substance, because only in these circumstances all the curves for the inner equilibria can run to the A-side with rise of temperature.

Yet this figure will not appear to be quite correct either, in my opinion, as a supposition is implied in it, which is highly improbable.

When B is a polymer of A, and the pseudo-binary system possesses a eutectic point, this means that there are liquids ($a\,c$) which contain more polymer than the coexisting solid phases (ad), and this is very improbable, so much so that we may disregard this figure altogether, in spite of HOLLMANN's [2]) assertion that he has found a eutectic point for the system acetaldehyde-paraldehyde. Probably this assertion of HOLLMANN's rests on not quite reliable observations, for my assistant, Mr. DE LEEUW, who tested the said assertion at my request, has not found it confirmed.

So for the case that B is a polymer of A and the two substances are *not* miscible in all proportions in the solid state, we must conclude to the existence of a Tx-figure as indicated by fig. 3, in which the

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1) In consequence of the considerable displacement of the inner equilibrium at the transition temperature it is possible, that while Q_r predominates *below* this temperature, *above* it the reverse takes place.

Q_r, too, can reverse its sign, but this seems less probable to me.

2) Zeitschr. f. phys. Chem. **43**, 129 (1903).

three-phase-temperature lies *between* the melting-points of the pseudo-components.

Now on this assumption, the solid phase possesses everywhere more of the polymer B than the coexisting liquid phase, and if in the unary system a transition point occurs, the course of the curves of inner equilibrium must be as indicated by kl_2, $S_2S'_2$ and $S_1'S'_3$.

If the curve kl_2 met the melting-point line of B, monotropy alone would be possible, as for enantiotropy reversal of the thermal sign would have to take place in this case, which is very improbable here.

Experimental confirmation.

It is clear that this theory requires that every substances which shows a transition point, must consist of two different kinds of molecules, which are in equilibrium at every temperature.

So if we consider the substance HgJ_2, the *red* modification of which passes into the *yellow* one at 127°, we must assume two different kinds of molecules, the former of which gives rise to the formation of *red*, and the other to that of *yellow* HgJ_2.

The investigation of this substance, which was carried out in collaboration with Mr. S. C. Bokhorst chem. cand. has led to a very remarkable result.

That it would appear that working quickly, the substance would betray its binary character, was expected, but that we should find here that case which I already mentioned, but considered as an exception, was highly surprising.

For the sake of clearness the observed phenomena will be discussed here in connection with the schematic fig. 4, in which α means yellow and β red HgJ_2.

At 127° the red phase passes into the yellow one, which new phase remains intensely yellow up to about 180°; on further heating we observed that this phase assumed a red colour, at first hardly perceptibly, but then more and more pronounced, and that it becomes a dark red liquid at the melting-point temperature 255°,4.

This phenomenon, which also appeared with very slow rise of the temperature, was studied in different ways with the naked eye and by means of the microscope, when it appeared that this change of colour takes place *continuously*, and is not owing to a second transition-point.

This continuous change of colour between comparatively narrow limits of temperature made it therefore probable that above the point of transition the curve for the solid inner equilibria at first

runs vertically upwards, after which it bends sharply to the red side, and meets the mixed-crystal curve of the pseudo-binary system near the axis of the red modification.

As therefore, this inner equilibrium curve appears to traverse the T-x-figure over a large concentration range, this pointed already to a region of partial-miscibility in the pseudo-binary figure, which was closed at the top, and so also to a continuous mixed crystal curve *acb*.

In order to test this supposition more closely, the following experiments were made with HgJ_2, either in thin-walled narrow capillaries or in so-called alcaloid tubes; it was, namely, quite immaterial which of these were taken, for in either case the experiment yielded the same result.

In these tubes the HgJ_2 was heated in a melting-apparatus up to a certain temperature *above* the transition-point, and then all at once transferred to an oil-bath of lower temperature, but always *above* the transition point.

The considerations which led us to these experiments, were the following.

If it is possible to make the cooling take place so rapidly that the inner equilibrium cannot keep pace with the temperature, the pseudo-binary character must appear, and entering the region of partial-miscibility the substance must split up into two phases.

Suppose that we start from the inner equilibrium p and that we cool this suddenly, in which not the curve of equilibrium, but the curve pS_3 is followed; then the red phase S_4 will appear by the side of the yellow phase S_3 and will have to be clearly visible.

This three-phase system will be strongly metastable, so that it is not to be expected that it will be very permanent; on the contrary, we may confidently predict that this state will very soon change into the only stable equilibrium which must lie on the curve SS_1.

If we now start from the inner equilibrium q, which lies on the right of the critical mixing-point K, the mixing-curve can be reached in S_5, and by the side of phase S_5, the phase S_6 must be found, which has a lighter colour.

As appears from the subjoined table (p. 770) not only these phenomena could be observed with great clearness, but moreover it was ascertained by these preliminary experiments that the mixing-point K must lie above 147° [1]).

Though it follows from these experiments that above the transition temperature the T,x-figure of the system HgJ_2 would be as indicated

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[1]) This investigation is continued in different directions.

Temp. HgJ_2	Suddenly cooled down to the temp.	Remarks
200°	130°	No unmixing as yet.
205°	"	Unmixing, *red* phase appears, but has disappeared again after a few seconds and the whole mass is again yellow.
207°	"	"
210°	"	"
212°	"	"
215°	"	"
225°	"	Unmixing, but now *yellow* phase appears and after a few seconds everything is yellow.
230°	"	The same phenomenon, and still more pronounced.
. . . . .	. . . . .	. . . . .
212°	140°	Unmixing *red* phase appears etc.
212°	145°	"
212°	147°	No unmixing is to be observed.

here, the question what the rest of the figure, i.e. under the transition point, would look like, remained unanswered. The answer to this question cannot yet be given in this communication, because the equilibrium sets in exceedingly slowly at temperatures under 100°.[1])

So the dotted curves under the transition temperature do not represent anything but a supposition. For the end in view here, however, the want of certainty *below* the transition temperature is of minor importance, as the phenomena observed at higher temperatures furnish a convincing proof for the validity of the theory.

Before I leave the substance HgJ_2 and proceed to another subject, I will only point out, that if the equilibria are considered not at constant pressure, but at the variable vapour-pressure, also the vapour-curves should be inserted in the T,x-figure, which lie on the side

[1]) If a tube with red HgJ_2 is immerged in liquid air, the colour becomes indeed much lighter viz. orange, but this change of colour has nothing to do with a displacement of the equilibrium.

If a mixture of *yellow* and *red* HgJ_2 is taken, and this is cooled down to —190°, the yellow colour changes into white, and the red into orange-yellow. When heated to the temperature of the room the heterogeneous mass is found to be entirely unchanged compared with the initial state.

of yellow HgJ_2, because the *yellow* phase is always deposited from the vapour.

If we now consider the question whether the literature mentions results in support of this theory, the answer is affirmative. These are chiefly the results obtained in the investigation of *sulphur* [1]) and that of *phosphorus* [2]).

In the system sulphur we have two different crystalline modifications, and besides them a third modification *Sμ*, which has not yet been obtained in crystalline form.

Considered in the light of this theory we must therefore assume *three* different kinds of molecules, and *sulphur* being known as a substance which is very slow, we can assume with great probability that *sulphur* is not pseudo-binary, but pseudo-ternary, i.e. will behave as a ternary system.

This, however, be only remarked in passing, as these considerations are of no further importance for what follows.

If we now direct our attention to the T,x-figure of the system *Sμ* and rhombic sulphur S_R (Fig. 5), it is noteworthy that by extrapolation **110°,6** has been found for the unary melting-point, and **112°,8** for the melting-point of pure rhombic sulphur.

It further appeared, however, that when from rhombic *S* was started from, where the equilibrium had set in at 90°, a melting-point was found at **110°.9**, the melting-point amounting to **111°.4** when the inner equilibrium had set in at ± 65°.

These are results which support the theory given here, for they point to the fact that we have to do here with a curve SS_1 for the solid inner equilibrium, which runs to the left with rise of temperature. For this curve shows that as we, working quickly, start from an inner equilibrium established at *lower* temperature, this phase will begin to melt at a *higher* temperature, which was also observed here.

The curve for the inner liquid equilibrium, too, runs to the left, so that the two curves of equilibrium have the same direction.

Though the sulphur can furnish further proofs, we now proceed to the phosphorus.

As Cohen and Olie already mentioned, investigations of Troost and Hautefeuille, Lemoine, Hittorf, and themselves point to the

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[1]) Kruyt, Z. f. phys. Chem. **64**, 513 (1908).

[2]) Cohen and Olie, Chem. weekblad **6**, 821 (1909).

fact that for phosphorus we have to do with solid inner equilibria between white and violet phosphorus.

If we consider the following results of the determinations of the specific gravity:

spec. grav. of red P obtained at $550° = 2{,}25$
" " " " " " " $450° = 2{,}28$
" " " " " " " $357° = 2{,}22$
" " " " " " " $255° = 2{,}20$
" " " " " " " $215° = 2{,}19$

we should, in view of the fact that the spec. grav. of white $P = 1{,}82$, and that of violet P may be put at about 2,34, come to the conclusion that the curve for the inner solid equilibria runs to the violet side with rise of temperature to 450°.

As it, however, followed from the experiments of COHEN and OLIE, that when red P was reduced from a higher to a lower temperature, the spec. grav. in general was *not* lowered, it is clear that they have not investigated states of equilibrium, and that we, therefore, cannot draw conclusions about the course of the curve of the inner equilibria from the above results. As to the existence of the inner equilibria, however, this is no longer doubtful.

So if we start from this, and if we then think of the phenomenon observed by CHAPMAN [1]) that red P when melting, gives a colourless liquid i.e. a liquid which perfectly resembles melted yellow P, a T, x-figure may be constructed in main lines for the pseudo-binary and the unary system, in which, however, the existence of a eutectic point is still an open question.

It has been assumed in fig. 6 that red $P(\beta P)$ is a polymer of white $P(\alpha P)$, and therefore no eutectic point is drawn. In this figure the phenomenon observed by CHAPMAN has been illustrated, for heated to the melting-point, the red solid phase will pass into a liquid l_2, which lies entirely on the side of the white P. We see further from this diagram that melted yellow P has about the same composition as melted red P, and that melted yellow P means undercooled liquid red P.

Applications.

Besides the phenomena mentioned here there are others which seen in the light of this theory find a plausible explanation. I allude

[1]) Journ. Chem. Soc. **75**, 743 (1899).

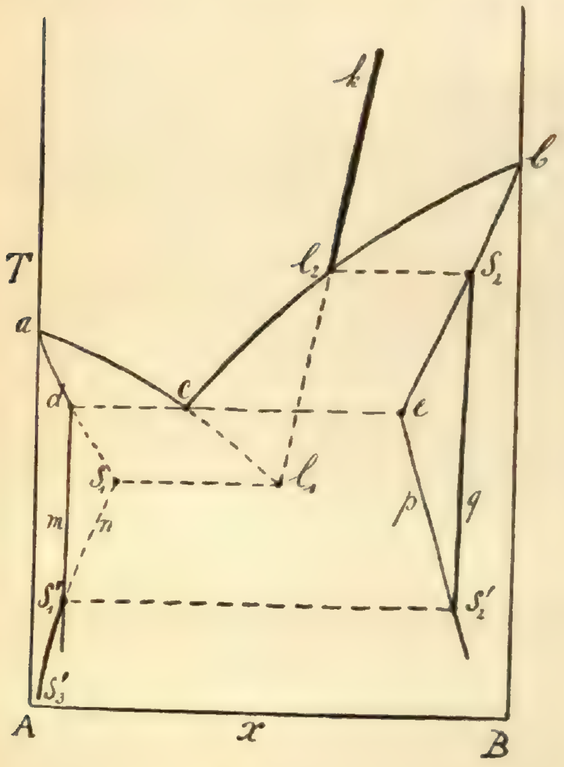

Fig. 1.

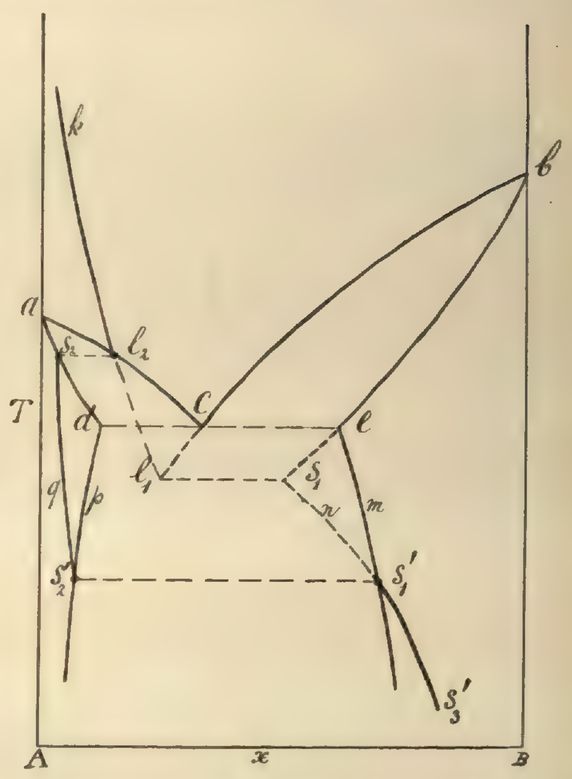

Fig. 2.

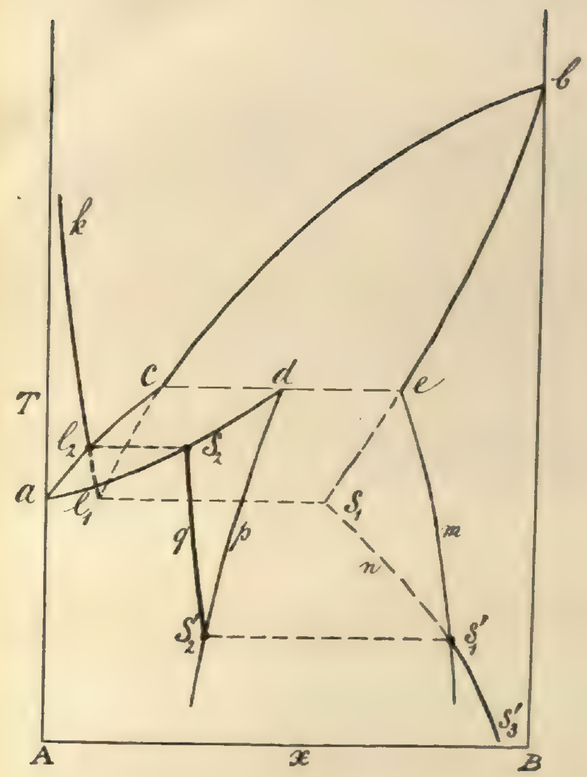

Fig. 3.

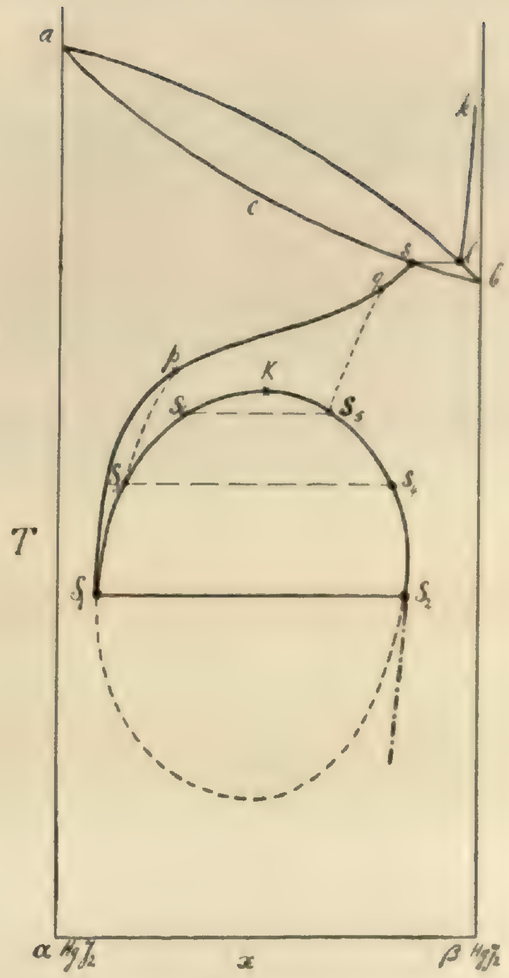

Fig. 4.

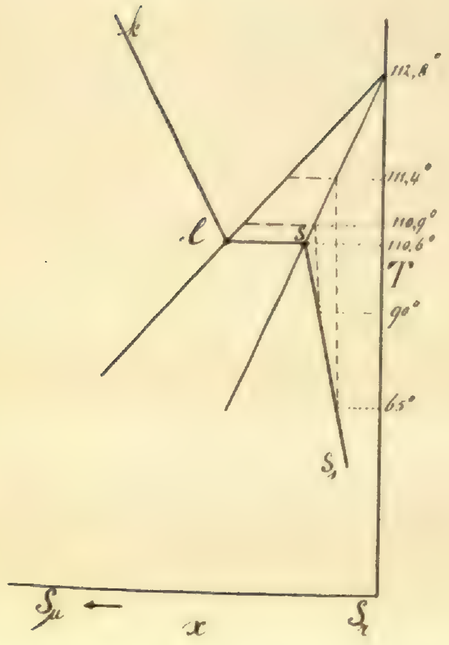

Fig. 5.

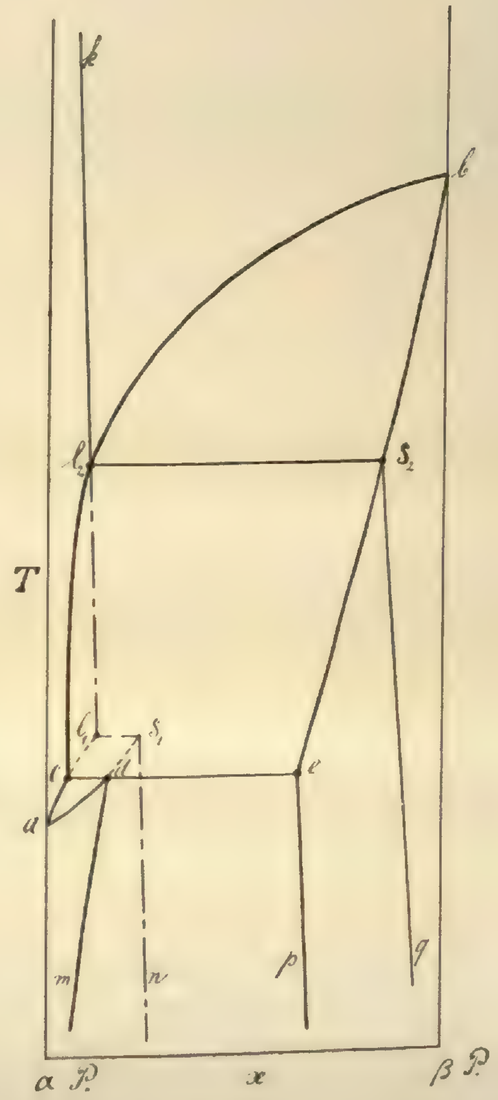

Fig. 6.

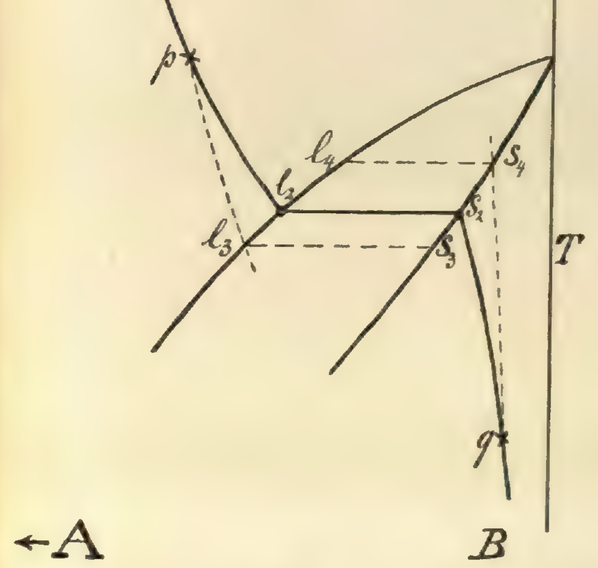

Fig. 7.

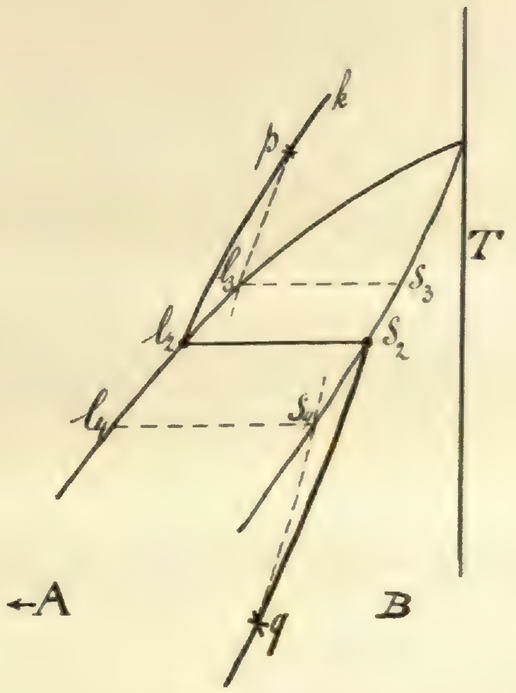

Fig. 8.

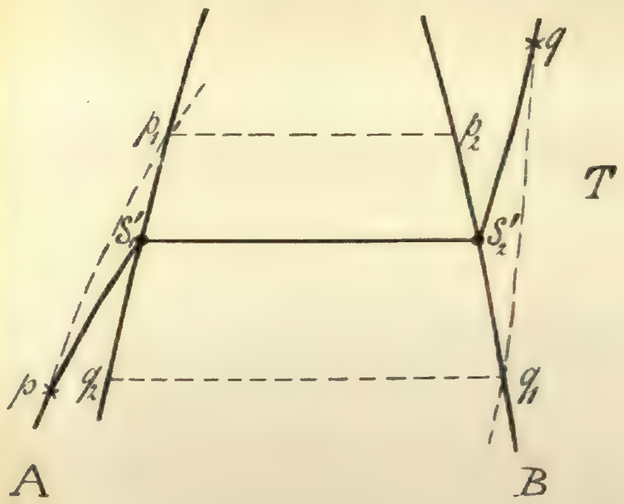

Fig. 9.

here to the phenomena of retardation *for so far they only appear when we work rapidly*[1]).

If we consider first of all the phenomenon of *undercooling* and *superheating* of the solid, for so far as they are only observed with quick change of temperature, fig. 7 gives a satisfactory explanation.

Starting from the inner liquid equilibrium p, not the curve of equilibrium pl_2, but another curve e.g. pl_3 will be followed with rapid cooling, and when we get beyond l_3 the state is not only unarily, but also pseudo-binarily *metastable*.

Let us assume for simplicity that in the pseudo-binary system no retardation worth mentioning appears, then the substance will solidify at l_3 and the solid substance S_3 is deposited.

Now this two-phase equilibrium is metastable to a high degree in the pseudo-binary system.

In the unary system equilibrium between liquid and solid substance can only exist under constant pressure at one temperature, and now it is the rule that a metastable state like that of the system $l_3 + S_3$ is at once destroyed. Thus we see e.g. that a supersaturate solution in contact with the substance which this solution must deposit to pass to the stable condition, generally immediately deposits this substance.

So the metastable two-phase equilibrium $l_3 + S_3$ is changed into the stable state $l_2 + S_2$, and this being a process which generates heat, the temperature rises to the unary melting-point.

Starting from the solid inner equilibrium q we get just the reverse, because then the substance melts at too high a temperature if quickly heated, as has already been observed for rhombic sulphur.

If now the curves of inner equilibrium run as in fig. 8, the liquid can solidify too early if cooled two rapidly, the solid substance can melt too early if heated too rapidly, and then the result is that for a *perfectly* pure substance there is a range of temperature over which the solidification and the melting extends, which probably often occurs for organic substances, in which the equilibrium sets in so slowly.

With regard to the phenomena of retardation at the transition point I need only refer to fig. 9, which will now be clear without further elucidation.

It is further hardly necessary to remark that when a substance is not *bi-*, but *tri-*, or *polymolecular*, the phenomena discussed here remain essentially the same.

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[1]) The peculiar phenomena, which will also appear for more complicated systems, as e.g. $Fe + C$ when we work quickly, will have to be accounted for in the same way.

In conclusion I want to point out that this theory gives the first plausible explanation of the *metastability of the metals.*

In this it is viz. noteworthy that the cooling of the solidified masses proceeds in such a way that the inner solid equilibrium can certainly not follow the temperature, and this is one of the reasons why the metals, as we generally have them, are nearly always in metastable state. We must further bear in mind that if we have a metal which is in inner equilibrium, and it is subjected to some mechanical operation, a necessary consequence of this will be that the metal becomes metastable, because in stable state a change of pressure is generally attended with a shifting of the inner equilibrium, which, however, in consequence of the inner resistance does not appear at all, or on account of the slight velocity of transformation will take place only after a very long time.

The above mentioned circumstances account at the same time for the fact that it hardly ever occurs that two pieces of the same metal are perfectly identical, for this could only occur when the inner state, stable or metastable, was perfectly the same.

Just as so many others the metastable states discussed here can be changed into the stable state by different influences, as increase of temperature, vibration, contact with the stable state etc., in which the transformation which takes place, manifests itself in a recrystallisation.[1])

Amsterdam, March 1910. *Anorg. Chem. Lab. of the University.*

ERRATA.

In the Proceedings of the Meetings of Jan. and Febr. 1910.

p. 652 line 9 and p. 677 line 5 from the bottom, p. 654 line 17 from the top: for 11 read 659.

p. 669, 672, 674 for 20.2 read 20.3.

p. 670 etc. for carrier read holder.
line 9 and 19 from the top: for modulus read constant.
line 5 from the bottom: for corresponding read in agreement with.

p. 672 line 16 from the bottom: for dilation read dilatation.

p. 673 for 14.3 read 14.0.

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[1]) It is to be expected that this metastability will not be met with only for metals and metal-alloys, but also for other substances, which have been obtained by rapid cooling and solidification of melted masses.

(April 28, 1910).

KONINKLIJKE AKADEMIE VAN WETENSCHAPPEN TE AMSTERDAM.

PROCEEDINGS OF THE MEETING

of Friday April 29, 1910.

(Translated from: Verslag van de gewone vergadering der Wis- en Natuurkundige Afdeeling van 29 April 1910, Dl. XVIII).

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Erratum, p. 845.

Mathematics. — "*On polar figures with respect to a plane cubic curve.* By Prof. JAN DE VRIES.

(Communicated in the meeting of March 26, 1910).

1. If a plane cubic curve γ^3 is represented symbolically by $a_x^3=0$ then $a_x\,a_y\,a_w=0$ represents the polar line p_{xy} of the points X and Y, i.e. the polar line of X with respect to the polar conic π_y of Y and at the same time the polar line of Y with respect to the polar conic π_x of X.

The three polar lines p_{xy}, p_{xz}, and p_{yz} will concur in one point W when the three conditions are satisfied

$$a_x a_y a_w = 0, \qquad a_x a_z a_w = 0, \qquad a_y a_z a_w = 0 \quad . \quad . \quad . \quad . \quad (1)$$

By elimination of the coordinates w_k we find out of it

$$(abc)\, a_x a_y b_x b_z c_y c_z = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

So to two given points X, Y belongs a conic γ_{xy}^2 as locus of the point Z; it passes also through X and Y, for when Z and X coincide, we find

$$(abc)\, a_x a_y b_x^2 c_y c_x \equiv (cba)\, c_x c_y b_x^2 a_y a_x \equiv -\,(abc)\, a_x a_y b_x^2 c_y c_x \equiv 0.$$

As we can substitute $(abc)\, a_x\, a_y\, c_x\, c_z\, b_y\, b_z = 0$ for (2), thus also $(abc)\, a_x\, a_y\, b_z\, c_z\, (b_x\, c_y - b_y\, c_x) = 0$, we can also represent γ_{xy}^2 by $(abc)\, a_x a_y b_z c_z\, (bc\xi) = 0$, where ξ_k are the coordinates of the line XY. Consequently (2) can be replaced by

$$(bc\xi)\,(bc\eta)\, b_z c_z = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

From this ensues that the conic γ_{xy}^2 is the *poloconica* $\pi_{\xi\eta}$ of the lines ξ and η.

So the poloconica of two lines is the locus of the points Z which with relation to the points of intersection X, Y of this conic with one of the given lines are in such a position that the polar lines p_{xz} and p_{yz} concur on the other one of the given lines, which is then at the same time polar line of X and Y.

2. If Z and W are the points of intersection of $\pi_{\xi\eta}$ with η, it follows out of the symmetry of (3) in connection with the equations (1), that the four points X, Y, Z, W form a closed group, so that each side of the quadrangle determined by them is the polar line of the vertices not lying on it, therefore *a polar quadrangle* (REYE).

Out of our considerations ensues that a polar quadrangle is determined by two of its vertices, but also by two of its opposite sides. In the last case the vertices are determined by the poloconica of the given lines; in the former case we can use the poloconica belonging to the polar line of the given points and their connecting line.

Out of $a_x a_y a_w = 0$ and $a_x a_y a_z = 0$ follows

$$a_x a_y (\lambda a_w + \mu a_z) = 0.$$

Here λ and μ can be determined in such a way that $\lambda a_w + \mu a_z = a_u$ relates to the point of intersection U of XY with WZ.

As $a_u a_x^2 = 0$ indicates the polar conic π_u of U we find that X and Y according to the relation $a_x a_y a_u = 0$ lie harmonically with respect to π_u. In an analogous way ensues from $a_w a_z a_y = 0$ and $a_w a_z a_x = 0$ the relation $a_w a_z a_u = 0$, according to which W and Z are also separated harmonically by π_u.

But then also the points $T \equiv (XZ, YW)$ and $V \equiv (XW, YZ)$ are conjugated with respect to π_u, i.e. we have $a_u a_v a_t = 0$. Now U, V, T are the *diagonal points* of the complete quadrangle $XYZW$, so that it is proved that *the diagonal triangle of a polar quadrangle is always a polar triangle* [1]).

3. When the conic γ_{xy}^2 degenerates we can take for Z each point on the line XY. To trace for this the condition, we put $z_k = \lambda x_k + \mu y_k$; from (2) follows

$$(abc)\, a_x a_y b_x c_y (\lambda b_x + \mu b_y)(\lambda c_x + \mu c_y) = 0,$$

so

$$\lambda^2 (abc)\, a_x a_y b_x^2 c_x c_y + \lambda\mu\, (abc)\, a_x a_y b_x^2 c_y^2 +$$

$$+ \lambda\mu\, (abc)\, a_x a_y b_x b_y c_x c_y + \mu^2\, (abc) a_x a_y b_x b_y c_y^2 = 0.$$

By exchanging two of the symbolic factors a, b, c, we see that three of these terms are identically zero; so we have

$$\lambda\mu\, (abc)\, a_x a_y b_x^2 c_y^2 = 0.$$

For an arbitrary choice of X and Y this equation furnishes only $\lambda = 0$ and $\mu = 0$, thus the points X and Y. It furnishes *each* point of XY, as soon as

$$(abc)\, a_x a_y b_x^2 c_y^2 = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

When X, Y, and Z are collinear, the polar line p_x of X and the polar lines p_{xy}, p_{xz} concur in *one* point; for these three lines are the polar lines of X, Y, Z with respect to the polar conic π_x. If now (4) is satisfied, then also p_{yz} passes through that point, hence, the six polar lines $p_x, p_y, p_z, p_{xy}, p_{yz}, p_{zx}$ concur in a point W. But when p_x, p_y and p_z are concurrent, the poloconica of $\xi \equiv XYZ$, degenerates and ξ is tangent of the *Cayleyana*.

From this ensues that for given Y the equation (4) will represent

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[1]) Mentioned without proof by Caporali (Transunti d. R. A. dei Lincei 1877, p. 236).

three right lines, namely the three tangents which we can draw out of Y to the *Cayleyana*.

This can be confirmed as follows. Let Z be a point of the locus of X, which is determined by (4) and X a second point of that locus lying on YZ, so that we have $a_x = \lambda a_y + \mu a_z$. Out of (4) then follows

$$(abc)\, a_y c_y^2 (\lambda a_y + \mu a_z)(\lambda b_y + \mu b_z)^2 = 0.$$

By exchanging a and c we see at once that

$$(abc)\, a_y^2 c_y^2 (\lambda b_y + \mu b_z)^2$$

vanishes identically. Analogously we find that $(abc)\, a_y c_y^2 a_z b_y^2$ and $(abc)\, a_y c_y^2 a_z b_y b_z$ vanish identically. As finally the form $(abc)\, a_y c_y^2 a_z b_z^2$ is zero because Z lies on the locus indicated by (4) the above relation is satisfied by all points of YZ, so the locus consists of three lines through Y.

4. That the line $\xi \equiv XY$ is tangent to the *Cayleyana* as soon as (4) is satisfied, can be confirmed by reducing (4) to the tangential equation of that curve. In the first place we find out of

$$(abc)\, a_x a_y b_x^2 c_y^2 = 0 \quad \text{and} \quad (acb)\, a_x a_y c_x^2 b_y^2 = 0$$

the relation

$$(abc)\, a_x a_y (b_x c_y + b_y c_x)(b_x c_y - b_y c_x) = 0.$$

The last factor can be replaced by $(bc\xi)$ where ξ_k indicate the coordinates of XY. After that the equation can be broken up into two terms, which pass into each other when b and c are exchanged. So we can replace it by

$$(abc)\, a_x a_y b_x c_y (bc\xi) = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

farthermore it is evident from

$$(abc)\, a_x a_y b_x^2 c_y^2 = 0 \quad \text{and} \quad (cba)\, c_x c_y b_x^2 a_y^2 = 0,$$

that at the same time is satisfied

$$(abc)\, b_x^2 a_y c_y (ac\xi) = 0,$$

so also

$$(bac)\, a_x^2 b_y c_y (bc\xi) = 0. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

By combining (5) and (6) we find

$$(abc)\, a_x c_y (bc\xi)(ab\xi) = 0.$$

So

$$(abc)\, c_x a_y (bc\xi)(ab\xi) = 0.$$

Out of the last two relations follows finally

$$(abc)\,(ac\xi)\,(bc\xi)\,(ab\xi) = 0. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

This tangential equation really represents the *Cayleyana* [1]).

So we have found that *the six polar lines* $p_x, p_y, p_z, p_{xy}, p_{yz}, p_{zx}$ *concur in one point when the points* X, Y, Z *lie on a tangent of the Cayleyana.*

5. When p_x, p_y, p_z are concurrent we have

$$(abc)\,a_x^2\,b_y^2\,c_z^2 = 0. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

This equation gives thus the relation between the coordinates of three points lying on one and the same polar conic.

For an arbitrary choice of X and Y this equation is satisfied except by X and Y by no point of the line XY. If it is to be satisfied by $z_k = \lambda x_k + \mu y_k$ we must have

$$(abc)\,a_x^2\,b_y^2\,(\lambda c_x + \mu c_y)^2 = 0,$$

therefore

$$\lambda\mu\,(abc)\,a_x^2\,b_y^2\,c_x\,c_y = 0.$$

This is satisfied for each value of $\lambda:\mu$ when the relation (4) is satisfied, so when X, Y, Z lie on a tangent of the *Cayleyana.*

Now in general the polar lines p_{xy}, p_{xz}, p_{yz} form a triangle inscribed in the triangle $p_x\,p_y\,p_z$ (see § 3). If (4) is satisfied then p_{xy}, p_{xz}, p_{yz} are concurrent; but then their point of intersection must be at the same time point of intersection of p_x, p_y, p_z.

If X, Y, Z are three collinear points of the cubic, then p_{yz}, p_{zx} and p_{xy} pass successively through X, Y, and Z.

For, from $a_x^3 = 0$, $a_y^3 = 0$ and $(\lambda a_x + \mu a_y)^3 = 0$ follows that the point Z is indicated by $a_x a_y\,(\lambda a_x + \mu a_y) = 0$. So we have $a_x a_y a_z = 0$, so Z lies on the polar line p_{xy}.

If moreover X, Y, Z lie on a tangent of the *Cayleyana*, then p_{yz}, p_{zx}, p_{xy} must coincide with the tangents p_x, p_y, p_z in X, Y, Z.

6. For p_x, p_y, and p_{xy} to be concurrent, there must be a point W for which we have $a_x^2 a_w = 0$, $b_y^2 b_w = 0$, and $c_x c_y c_w = 0$.

But then $(abc)\,a_x^2 b_y^2 c_x c_y = 0$.

For arbitrarily chosen Y the locus of X becomes a figure of the third order, passing through Y, because we have $(abc)\,a_y^2 b_y^2 c_y^2 = 0$. But by taking notice of (4) we see that this figure consists of three tangents of the *Cayleyana*. Out of

$$a_x^2 a_w = 0,\ a_y^2 a_w = 0 \text{ and } a_x a_y a_w = 0$$

[1]) See e.g. CLEBSCH, Leçons sur la géométrie, II, p. 284.

follows indeed

$$(\lambda a_x + \mu a_y)^2 a_w = 0;$$

i.e. if Z lies on XY, then p_z will pass through the point of intersection W of p_x, p_y and p_{xy}, which bears then at the same time p_{yz} and p_{xz}.

So the three lines p_x, p_y, and p_{xy} concur only then in one point when X and Y are united by a tangent of the *Cayleyana*. Their point of intersection bears then also all the polar lines and mixed polar lines belonging to the points of those lines.

The lines p_x, p_{xy}, and p_{xz} will be concurring, when

$$(abc)\, a_x^2 b_x b_y c_x c_z = 0$$

is satisfied, thus also

$$(abc)\, a_x^2 c_x c_y b_x b_z = 0,$$

hence also

$$(abc)\, a_x^2 b_x c_x\, (b_y c_z - b_z c_y) = 0.$$

If we put

$$y_k z_l - y_l z_k = \xi_m,$$

we have the condition

$$(abc)\, a_x^2 b_x c_x\, (bc\xi) = 0.$$

As this can also be written in the forms

$$(abc)\, a_x b_x^2 c_x\, (ac\xi) = 0 \quad \text{and} \quad (abc)\, a_x b_x c_x^2\, (ab\xi) = 0$$

and as out of

$$\begin{vmatrix} a_1 & a_2 & a_3 & a_x \\ b_1 & b_2 & b_3 & b_x \\ c_1 & c_2 & c_3 & c_x \\ \xi_1 & \xi_2 & \xi_3 & \xi_x \end{vmatrix} = 0$$

follows the relation

$$(abc)\, \xi_x = a_x\, (bc\xi) + b_x\, (ca\xi) + c_x\, (ab\xi)$$

the above condition can be replaced by

$$(abc)^2\, a_x b_x c_x \xi_x = 0.$$

With arbitrary position of X this is satisfied by $\xi_x = 0$, i.e. when X, Y, and Z are collinear (see § 3).

If however

$$(abc)^2\, a_x b_x c_x = 0,$$

so that X lies on the *Hessian*, then X, Y, and Z are quite arbitrary. This was to be foreseen, now namely π_x is a pair of lines, so that the lines p_x, p_{xy}, and p_{xz} concur in the node of π_x.

Physiology. — *"Contributions to the study of serum-anaphylaxis."* (From the "Institut für Infektionskrankheiten" at Berlin). By Dr. J. G. SLEESWIJK. (Communicated by Prof. C. H. H. SPRONCK). (4th communication.)

(Communicated in the meeting of March 26, 1910).

During the first months of last year I had an opportunity, in three communications [1]), to make the results of my investigations about serumanaphylaxis known to the Academy. Since that time the literature about this subject has not inconsiderably increased. It is not my intention at all here to go in for a discussion of this. Only let it be allowed to me (and I even consider myself obliged to do this before the Academy) to treat here in a few words of these publications, in which my own investigations either directly or indirectly were discussed, and to add the results of a number of further experiments. May it be presupposed that in general the facts communicated by me must be acknowledged as correct.

Principally we have to pay attention to three points:

1st. the part of the red corpuscles of the guinea-pig with respect to horse-serum in the phenomenon of TH. SMITH, 2nd. the problem of the alexine-fixation and of the haemolysis in the anaphylactic shock, and 3rd the application of the specific hypersensibility for proteins in medicina forensis.

Last year I explained why I was of opinion that the sensitizing principle of the first injection and the toxic substance of the second administering of serum must be considered identical and that only quantitative differences are met with here. In the meantime BESREDKA [2]) has changed his mind and taken this standpoint. Now the logical consequence of my observation that horse-serum by treatment with the blood of a guinea-pig can be deprived of its poisonousness for animals made sensitive, was therefore that with this at the same time the sensitizing substance is fixed. LEVADITI and RAYCHMAN, who could also really prove what I mentioned last, do not refrain, therefore, in this connection, from referring to my communication. [3]) Also SALUS corroborated my observation concerning the depoisoning action of the red corpuscles on horse-serum. [4])

The problem of the complement fixation has of late attracted attention to a high degree. I remind of my first communication in which I said already, "that a sensitized guinea-pig, which reacts

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[1]) These Proceedings: January, February, and March 1909.
[2]) Ann. de l'Inst. Pasteur, Oct. 1909.
[3]) C. R. Soc. de Biol. T. 67, 1909, p. 1078.
[4]) Wiener Klin. Woch. 1909, no. 48.

upon the second serumadministration with symptoms of intoxication some time after that injection produces a serum that is exceedingly poor in haemolytic alexine." By the side of this I have proved that the serum of hypersensitive animals in not a single combination with horse-serum gives a precipitate, nor is it able to fix complement. A remarkable incongruity therefore, and by which the anaphylactic state is distinguished from the phase of immunity, at which praecipitines are formed, and at which in vivo as well as in the test-tube alexine is fixed. I point out that here I came in conflict with NICOLLE and ABT [1]), who had found that the serum of sensitized guinea-pigs does fix complement with horse-serum in vitro. For FRIEDBERGER, who considers anaphylaxis as a peculiar form of the immunity for proteins, at which the praecipitines only for a trifling part have passed into circulation, but principally have remained fixed (sessile) to the cells, the fixation of complement was a welcome phenomenon. He took for granted (evidently without any further control) that the communication of NICOLLE and ABT was correct, whilst, on the other hand, he could confirm by his own investigation my observation about the loss of alexine during the anaphylactic shock. [2]) Yet even here such quantitative differences came to light that at first sight my observations seemed to show a shade of incorrectness. I had said for example, that the maximum of complement-loss is reached after about half an hour, whereas FRIEDBERGER found this to be the case already within five minutes. Therefore I am compelled to enter into this somewhat more closeiy.

FRIEDBERGER has evidently not asked himself where the cause may lie of our diverging results, quantitative as they may only be. He speaks only in passing of "Differenzen in der angewandten Anaphylaxie-technik." But here lies the cardo quaestionis, and what I presumed already, appeared to me on closer investigation to be reality. I had namely administered to my animals the toxic serum-injection in not too great a dose in their abdomen; the reaction is then less violent and has a slower course, so that — through the investigation of blood-samples taken from the animals consecutively at different times of the anaphylactic shock — I could in general fix the course of the complement-curve. FRIEDBERGER, however, injected. his hypersensitive animals intravenously: the reaction then goes so quickly and is so violent, that the guinea-pigs usually die within few minutes. And at the same time also the complement-loss has soon reached its maximum. Now in WASSERMANN's laboratory I have

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1) Ann. de l'Inst. Pasteur 1908.

2) Zeitschr. f. Imm. forsch. Bd. III, H. 6.

been able to fix these differences in a series of exact quantitative complement-titrations. From these it appeared among others that a few minutes after intravenous injection of 0.2 cm³ of horse-serum the complement-quantity of the testanimal-serum may have decreased nearly as strongly as at intraperitoneal injection of 3 cm³ after half an hour.

My investigations and those of FRIEDBERGER, accordingly, do not contradict each other; they complete each other. Therefore I cannot see in FRIEDBERGER's results anything but an essential corroboration of my observations. The same thing holds good for the haemolysis, which in the anaphylactic shock shows itself in the test-animals. FRIEDBERGER corroborates also this fact, just as POELS [1]) does. My contention, therefore, that BESREDKA with his exclusivistic opinion that it is only the elements of the central nervous system which are to be brought to hypersensibility, is wrong, finds satisfactory support in this haemolysis which has been proved in more than one hand.

Now as to the alexine-fixation of the anaphylactic serum with its antigen in vitro, I think I can maintain my negative results over-against NICOLLE and ABT. In many series with mixtures of falling quantities of horse-serum with rising quantities of anaphylactic guineapig-serum I could not observe anywhere a specific retardment of the haemolysis. Not even though I stuck accurately to the quantitative proportions, as NICOLLE and ABT have mentioned. In the meantime, thanks to the necessary controlling experiments, I came to the following conclusions. Even normal guinea-pig serum (inactivated) often retard the haemolysis, quite independent of the presence of horse-serum; nay, we sometimes meet a normal serum which has a stronger fixing power than an anaphylactic serum which has also been examined [2]). Therefore I abide by my former contention that there is here no question of a specific complement-fixation in the test-tube. This incongruity of the alexine-fixation in vitro and in vivo, to which I drew attention already a year ago, was the other day corroborated with certainty by MICHAËLIS in the meeting of the "Physiologische Gesellschaft" at Berlin (21 January 1910). Also FRIEDBERGER seems after all to share this opinion (Zeitschr. f. Imm. forsch. Bd. IV, H. 5). It now seems to me that the labile state of physiological equilibrium, in which the hypersensitive organism finds itself, is biologically characterized by the incongruity referred to just now.

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[1]) Handelingen v. h. Nederl. Natuur- en Geneesk. Congr. te Utrecht, April 1909.

[2]) Ampler details I hope to publish elsewhere.

Not long ago TSURUN [2]) tried to reduce the signification of the alexine fixation in vivo in anaphylaxis. Thus already in normal guinea-pigs normal serum from dog or rabbit would cause loss of complement. This certainly does not hold good for horse serum, as had already appeared from my former controlling experiments; indeed, about this TSURUN does not speak. But moreover this investigator has worked with corpuscles sensitized not strongly enough and with insufficient dilutions of complement, so that his results do not deserve a very great confidence. Leaving this out of consideration, he found, just as I did before, that the intoxication-phenomena and the loss of complement need not run parallel, from which I drew the conclusion that these two are not directly dependent upon each other, but have a common cause.

I now come to the third point that I wish to treat of here, viz. the application of anaphylaxis in the practice of medicina forensis. Evidently this application was so clear that about at the same time and independent of my communication, similar results were made known by THOMSEN, UHLENHUTH, and H. PFEIFFER. The last lays stress upon the strong fall in the temperature of the body during the anaphylactic shock as a resource for the diagnosis.

Concerning the technique of this investigation the following may still be mentioned. If a blood spot has to be identified, and if guinea-pigs are treated intraperitoneally, several cm^3 of serum are necessary for each animal that is to be examined. If the animals are treated intravenously (in the jugularis), much smaller quantities of serum are wanted, but then an operation in the neck has to take place, which, however, after some practice for this purpose offers no objection. It has now appeared to me that also with young rabbits of about 1 K.G. the experiment can be very well made, because here both the sensitizing and the trying injection can be easily made in the earvena. A small dried up blood-spot is dissolved in 1 cm^3. of physiological salt-solution and injected in such an animal; after a fortnight 1 cm^3 of the suspected kinds of blood is injected also intravenously. To rabbits, which had thus been previously treated with extracts from human blood-spots, I have administered on consecutive days serum from goat, horse, cow, and guinea-pig, without the animals reacting in the least. Lastly 1 cm^3 of human serum caused them within a few minutes to answer with spasms and paralyses, with respiratory disturbances, incontinentia urinae et alvi, etc. The anaphylactic reaction will, in my opinion, in the practice

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[1]) Zeitschr. f. Imm. forsch Bd. IV, H. 5.

of medicina forensis henceforth maintain its position by the side of the precipitation in vitro as a valuable method.

Among the many questions that show themselves in the study of our subject, there was also the following: what happens to the injected horse-serum during the anaphylactic shock? If there were only a minimal quantity free and unchanged in the circulation of the intoxicated animal, it ought to be possible that with its blood a normal animal could be sensitized. Now it has appeared to me that this is never crowned with success. From this it may be inferred that all the antigen taken up in the blood-circulation is at once fixed by the cells of the hypersensitive organism, resp. deprived of its specific character at the same time.

Mathematics. — "*On the structure of perfect sets of points*". By Dr. L. E. J. Brouwer. (Communicated by Prof. Korteweg).

(Communicated in the meeting of March 26, 1909).

§ 1.

Sets of points and sets of pieces.

The sets of points discussed in the following lines are supposed to be lying within a finite domain of a Sp_n.

By a *piece* of a closed set of points μ we understand a *single point or closed coherent set of points*, belonging to μ, and not contained in an other closed coherent set of points belonging to μ.

We can regard as elements of μ its pieces as well as its points, in other words we can consider μ on one hand as a *set of points*, on the other hand as a *set of pieces*.

Let us choose among the pieces of μ a fundamental series $S_1, S_2, S_3, \ldots$, then to μ belong one or more pieces ${}_1S_\omega$, ${}_2S_\omega, \ldots$ with the property that S_n lies entirely within a for indefinitely increasing n indefinitely decreasing distance ε_n from one of the pieces ${}_\alpha S_\omega$. These parts ${}_\alpha S_\omega$ we shall call the *limiting pieces* of the fundamental series $S_1, S_2, S_3, \ldots$

As thus the set μ possesses to each of its fundamental series of pieces at least one limiting piece, *a closed set of points is likewise closed as set of pieces*.

By an *isolated piece* of μ we understand a piece having from its rest set in μ a finite distance, in other words a piece, the rest set of which is closed.

THEOREM 1. *Each piece of μ is either a limiting piece, or an isolated piece.*

Let namely S be a non-isolated piece, then there exists in μ a fundamental series of points $t_1, t_2, t_3, \ldots$. not belonging to S, converging to a single point t of S. If t_1 lies on S_1, then S_1 has a certain distance ε_1 from S. There is then certainly a point t'_2 of the fundamental series possessing a distance $< \varepsilon_1$ from S, lying therefore not on S_1 but on an other piece S_2. Let ε_2 be the distance of S_2 from S, then there is certainly a point t'_3 of the fundamental series possessing a distance $< \varepsilon_2$ from S, lying thus neither on S_1, nor on S_2, but on a third piece S_3. Continuing in this manner we determine a fundamental series of pieces $S_1, S_2, S_3, \ldots$, containing consecutively the points $t_1, t'_2, t'_3, \ldots$ converging to t. So the pieces $S_1, S_2, S_3, \ldots$ converge to a single limiting piece which can be no other than S.

By a *perfect set of pieces* we understand a closed set, of which each piece is a limiting piece.

A perfect set of pieces is also perfect as set of points; but the inverse does not hold. For, a perfect set of points can very well contain isolated pieces.

We shall say that two sets of pieces *possess the same geometric type of order*, when they can be brought piece by piece into such a one-one correspondence, that to a limiting piece of a fundamental series in one set corresponds a limiting piece of the corresponding fundamental series in the other set. So in general a closed set considered as a set of pieces possesses not the same geometric type of order as when considered as a set of points.

A closed set we shall call *punctual*, when it does not contain a coherent part, in other words when all its pieces are points.

§ 2.

Cantor's fundamental theorem and its extensions.

The fundamental theorem of the theory of sets of points runs as follows:

If we destroy in a closed set an isolated point, in the rest set again an isolated point, and so on transfinitely, this process leads after a denumerable number of steps to an end.

The discoverers of this theorem, CANTOR [1]) and BENDIXSON [2]) proved

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[1]) Mathem. Annalen 23, p. 459—471.

[2]) Acta Mathematica 2, p. 419—427.

it with the aid of the notion of the *second transfinite cardinal* Ω, which is however not recognised by all mathematicians. LINDELÖF [1]) gave a proof independent of this notion, where, however, the process of destruction itself remaining non-considered, the result is more or less obtained by surprise.

Only for linear sets there have been given proofs of the fundamental theorem, which at the same time follow the process of destruction and are independent of Ω [2]).

The rest set which remains after completion of the process of destruction and which we may call the *Cantor residue*, is after CANTOR [3]) *a perfect set of points*, however of the most general kind, thus *in general not a perfect set of pieces.*

An extension of the fundamental theorem, enunciated by SCHOENFLIES[4]) and proved by me [5]), can be formulated as follows:

If we destroy in a closed set an isolated piece, in the rest set again an isolated piece, and so on transfinitely, this process leads after a denumerable number of steps to an end.

My proof given formerly for this theorem was a generalisation of LINDELÖF's method, but at the same time I announced a proof which follows the process of destruction, and which I give now here; in it is contained a proof of the fundamental theorem, which in simplicity surpasses by far the existing ones, is independent of Ω, and follows the process of destruction:

By means of Sp_{n-1}'s belonging to an orthogonal system of directions we divide the Sp_n into n-dimensional cubes with edge a, each of these cubes into 2^n cubes with edge $\frac{1}{2}a$, each of the latter into 2^n cubes with edge $\frac{1}{4}a$, etc.

All cubes constructed in this way form together a denumerable set of cubes K.

Let now μ be the given closed set, then K possesses as a part a likewise denumerable set K_1 consisting of those cubes which contain in their interior or on their boundary points of μ.

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[1]) Acta Mathematica 29, p. 183—190.

[2]) SCHOENFLIES, Bericht über die Mengenlehre I, p. 80, 81; Gött. Nachr. 1903, p. 21—31; HARDY, Mess. of Mathematics 33, p. 67—69; YOUNG, Proceedings of the London Math. Soc. (2) 1, p. 230—246.

[3]) l. c. p. 465.

[4]) Mathem. Annalen 59; the proof given there p. 141—145, and Bericht über die Mengenlehre II, p. 131—135 does not hold.

[5]) Mathem. Annalen 68, p. 429.

To each destruction of an isolated point or isolated piece in μ now answers a destruction of at least *one* [4]) cube in K_1; but of the latter destructions only a denumerable number is possible, thus also of the former, with which CANTOR's theorem and SCHOENFLIES's theorem are proved both together.

Let us call the rest set, which remains after destruction of all isolable pieces, the *Schoenflies residue*, then on the ground of theorem 1 we can formulate:

THEOREM 2. *A Schoenflies residue is a perfect set of pieces.*

§ 3.

The structure of perfect sets of pieces.

Let S_1 and S_2 be two pieces of a perfect set of pieces μ. Let it be possible to place a finite number of pieces of μ into a row having S_1 as its first element and S_2 as its last element in such a way, that the distance between two consecutive pieces of that row *is smaller than* a. Then we say, *that* S_2 *belongs to the* a*-group of* S_1.

If S_2 and S_3 both belong to the a-group of S_1, then S_3 belongs also to the a-group of S_2, so that μ breaks up into a certain number of "a-groups". This number is finite, because the distance of two different a-groups cannot be smaller than a.

If $a_1 < a_2$, and if an a_1-group and an a_2-group of μ are given, then these are either entirely separated or the a_1-group is contained in the a_2-group.

If two pieces S_1 and S_2 of μ are given, then there is a certain maximum value of a, for which S_1 and S_2 lie in different a-groups of μ. That value we shall call the *separating bound of* S_1 *and* S_2 *in* μ, and we shall represent it by $\sigma_\mu(S_1, S_2)$.

If fartheron we represent the *distance of* S_1 *and* S_2 by $\alpha(S_1, S_2)$, then $\sigma_\mu(S_1, S_2)$ converges with $\alpha(S_1, S_2)$ to zero, but also inversely $\alpha(S_1, S_2)$ with $\sigma_\mu(S_1, S_2)$. For otherwise convergency of $\sigma_\mu(S_1, S_2)$ to zero would involve the existence of a coherent part of μ, in which two different pieces of μ were contained, which is impossible.

The maximum value of a for which μ breaks up into different a-groups we shall call the *width of dispersion* of μ, and shall represent it by $\delta(\mu)$. This width of dispersion of μ is at the same time the greatest value which $\sigma_\mu(S_1, S_2)$ can reach for two pieces S_1 and S_2 of μ.

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[4]) Even of an infinite number.

The maximum value of a, for which μ breaks up into *at least* n different a-groups we shall call the *n-partite width of dispersion* of μ, and shall represent it by $\delta_n(\mu)$. Clearly $\delta_n(\mu)$ is $\leqq \delta(\mu)$.

For μ exists furthermore a series of increasing positive integers $n_1(\mu)$, $n_2(\mu)$, $n_3(\mu)$, ... in such a way that $\delta_n(\mu)$ for n between $n_{k-1}(\mu)$ and $n_k(\mu)$ is equal to $\delta_{n_k(\mu)}(\mu)$. This quantity $\delta_{n_k(\mu)}(\mu)$ we call the k^{th} *width of dispersion* of μ and as such we represent it by $\delta^{(k)}(\mu)$.

We now assert that it is always possible to break up μ into m_1 perfect sets of pieces $\mu_1, \ldots . \mu_{m_1}$ so as to have $\delta(\mu_h) \leqq \delta_{m_1}(\mu)$ and $\alpha(\mu_{h_1}, \mu_{h_2}) \geqq \delta_{m_1}(\mu)$.

Let namely be $\delta_{m_1}(\mu) = \delta^{(k)}(\mu)$; we can then obtain the required number m_1 by composing each μ_h of a certain number of $\delta^{(k)}(\mu)$-groups belonging to a same $\delta^{(k-1)}(\mu)$-group. We are then also sure of having satisfied the condition $\alpha(\mu_{h_1}, \mu_{h_2}) \geqq \delta_{m_1}(\mu)$.

Fartheron we can place the $\delta^{(k)}(\mu)$-groups of a same $\delta^{(k-1)}(\mu)$-group into such a row that the distance between two consecutive ones is equal to $\delta^{(k)}(\mu)$. If we take care that each μ_h consists of a non-interrupted segment of such a row, then the condition $\delta(\mu_h) \leqq \delta_{m_1}(\mu)$ is also satisfied.

Let us now break up in the same way each μ_h into m_2 perfect sets of pieces $\mu_{h1}, \ldots . \mu_{hm_2}$ in such a way that $\delta(\mu_{hi}) \leqq \delta_{m_2}(\mu_h)$ and $\alpha(\mu_{hi_1}, \mu_{hi_2}) \geqq \delta_{m_2}(\mu_h)$, and let us continue this process indefinitely.

If then we represent by F_ν an arbitrary row of ν indices, then we shall always find

$$\delta_{m_\nu}(\mu_{F_{\nu-1}}) \leqq \delta_{m_1+m_2+\ldots+m_\nu+1-\nu}(\mu) \quad . \quad . \quad . \quad . \quad (A)$$

As μ is a perfect set of pieces, the width of dispersion $\delta(\mu_{F_\nu})$ can converge to zero only for indefinite increase of ν; out of the formula (A) follows, however, that for indefinite increase of ν that convergency to zero always takes place and, indeed, uniformly for all ν^{th} elements of decomposition together.

At the same time the separating bound of every two pieces lying in one and the same ν^{th} element of decomposition converges uniformly to zero; so these elements of decomposition converge themselves uniformly each to a single piece.

If finally a variable pair of pieces of μ is given, then their distance can converge to zero only when the order of the smallest element of decomposition, in which both are contained, increases indefinitely.

The simplest mode in which this process of decomposition can be

executed is by taking all m_k's equal to 2. If then we represent the two elements of decomposition of the first order by μ_0 and μ_2, those of the second order by $\mu_{00}, \mu_{02}, \mu_{20}, \mu_{22}$, and so on, then in this way the different pieces of μ are brought into a one-one correspondence with the different fundamental series consisting of figures 0 and 2. And two pieces converge to each other then, and only then, when the commencing segment which is common to their fundamental series, increases indefinitely.

Let us consider on the other hand, in the linear continuum of real numbers between 0 and 1, the perfect punctual set π of those numbers which can be represented in the triadic system by an infinite number of figures 0 and 2. The geometric type of order of π we shall represent by ξ.

Two numbers of π converge to each other then and only then, when the commencing segment which is common to their series of figures, increases indefinitely.

So, if we realize such a *one-one* correspondence between the pieces of μ and the numbers of π, that for each piece of μ the series of indices is equal to the series of figures of the corresponding number of π, then to a limiting piece of a fundamental series of pieces of μ corresponds a limiting number of the corresponding series of numbers in π, so that we can formulate:

THEOREM 3. *Each perfect set of pieces possesses the geometric type of order* ξ.

For the case that the set under discussion *is punctual and lies in a plane,* this theorem ensues immediately from the following well-known property:

Through each plane closed punctual set we can bring an arc of simple curve.

Combining SCHOENFLIES's theorem mentioned in § 2 with theorem 3 we can say:

THEOREM 4. *Each closed set consists of two sets of pieces; one of them possesses, if it does not vanish, the geometric type of order* ξ, *and the other is denumerable.*

§ 4.

The groups which transform the geometric type of order ξ *in itself.*

Just as spaces admit of groups of continuous one-one transformations, whose geometric types of order [1]) are again spaces, namely

[1]) In this special case formerly called by me "Parametermannigfaltigkeiten" Comp. Mathem. Annalen 67, p. 247.

the finite continuous groups of LIE, the geometric type of order ζ admits of groups of continuous one-one transformations, which possess likewise the geometric type of order ζ.

In order to construct such groups we start from a decomposition according to § 3 of the set μ into m_1 "parts of the first order" $\mu_1, \mu_2, \ldots \mu_{m_1}$, of each of these parts of the first order into m_2 "parts of the second order" $\mu_{h1}, \mu_{h2}, \mu_{h3}, \ldots \mu_{hm_2}$, etc.

The parts of the first order we submit to an arbitrary transitive substitution group of m_1 elements, of which we represent the order by p_1, and which we represent itself by g_1.

After this we submit the parts of the second order to a transitive substitution group g_2 of $m_1 m_2$ elements which possesses the parts of the first order as systems of imprimitivity and g_1 as substitution group of those systems into each other. We can then represent the order of g_2 by $p_1 p_2$.

The simplest way to construct such a group g_2, is to choose it as the direct product of g_1 and a substitution group γ_2, which of the parts of the second order leaves the first index unchanged and transforms the second index according to a single transitive substitution group of m_2 elements.

We then submit the parts of the third order to a transitive substitution group g_3 of $m_1 m_2 m_3$ elements which possesses the parts of the second order as systems of imprimitivity and g_2 as substitution group of those systems into each other. We can represent the order of g_3 by $p_1 p_2 p_3$.

In this way we construct a fundamental series of substitution groups $g_1, g_2, g_3, \ldots$

Let τ_1 be an arbitrary substitution of g_1; τ_2 a substitution of g_2 having on the first index of the parts of the second order the same influence as τ_1; τ_3 a substitution of g_3 having on the first two indices of the parts of the third order the same influence as τ_2; and so on.

The whole of the substitutions τ_n then determines a substitution of the different fundamental series of indices into each other, in other words *a transformation τ of the pieces of μ into each other.*

This transformation is in the first place a *one-one transformation*; for, two different pieces of μ lie in two different parts of a certain, e.g. of the r^{th} order, and these are transformed by τ into again two different parts of the r^{th} order.

If fartheron $S_1, S_2, S_3, \ldots$ is a fundamental series of pieces, possessing S_ω as its only limiting piece, then, if $\lambda(n)$ is the lowest possible order with the property that S_n and S_ω lie in different parts of that order, $\lambda(n)$ must increase indefinitely with n.

So by the transformation τ the fundamental series passes into a new fundamental series having as its only limiting piece the piece into which S_ω passes by τ.

As a set of pieces μ is thus *continuously* transformed by τ.

Let $\tau'_1, \tau'_2, \tau'_3, \ldots$ be a series of substitutions satisfying the same conditions as the series $\tau_1, \tau_2, \tau_3, \ldots$ If then $\tau_1 \tau'_1 = \tau''_1$; $\tau_2 \tau'_2 = \tau''_2$; etc., then the series $\tau''_1, \tau''_2, \tau''_3, \ldots$ likewise satisfies the same conditions.

If farthermore τ' and τ'' are defined analogously to τ, then $\tau\tau'$ is equal to τ''.

So the transformations satisfying the conditions put for τ form a group, which we shall represent by g.

To investigate the geometric type of order of this group, we decompose in the way indicated in § 3 a perfect set of pieces ϱ into p_1 parts of the first order $\varrho_1, \varrho_2, \ldots, \varrho_{p_1}$; each of these into p_2 parts of the second order $\varrho_{h1}, \varrho_{h2}, \ldots, \varrho_{hp_2}$; and so on.

The p_1 substitutions of g_1 we bring into a one-one correspondence to the parts of the first order of ϱ. Then the $p_1 p_2$ substitutions of g_2 into such a one-one correspondence to the parts of the second order of ϱ, that, if a substitution of g_2 and a substitution of g_1 have the same influence on the first index of the parts of the second order of μ, the part of the second order of ϱ corresponding to the former lies in the part of the first order of ϱ corresponding to the latter.

In like manner we bring the $p_1\ p_2\ p_3$ substitutions of g_3 into such a one-one correspondence to the parts of the third order of ϱ, that, if a substitution of g_3 and a substitution of g_2 have the same influence on the first two indices of the parts of the third order of μ, the part of the third order of ϱ corresponding to the former lies in the part of the second order of ϱ corresponding to the latter; and so on.

The parts of ϱ corresponding to a series $\tau_1, \tau_2, \tau_3, \ldots$ then converge to a single piece of ϱ, which we let answer to the transformation τ deduced from the series. Then also inversely to each piece of ϱ answers a transformation τ, and the correspondenceattained in this manner is *a one-one correspondence.*

Farthermore two transformations τ and τ' converge to each other then and only then, when their generating series $\tau_1, \tau_2, \tau_3, \ldots$ and $\tau'_1, \tau'_2, \tau'_3, \ldots$ have an indefinitely increasing commencing segment in common, in other words when the corresponding pieces of ϱ converge to each other. So the correspondence between the transformations τ and the pieces of ϱ is *continuous.*

The transformations τ, in other words the transformations of the group g, have thus been brought into a continuous one-one correspondence to the pieces of ϱ, *so that g possesses the geometric type of order ζ.*

If now we adjoin to each substitution group g_n a finite group g'_n of continuous one-one transformations of μ as a set of pieces in itself, tranforming of the pieces of μ the first n indices according to g_n, but leaving unchanged all their other indices, *then the fundamental series of the groups* $g'_1, g'_2, g'_3, \ldots$ *converges uniformly to the group* g.

The set whose elements are the groups g of the geometric type of order ζ constructable in the indicated manner possesses the cardinal number of the continuum. For, already the set of those series $m_1, m_2, m_3, \ldots$, which consist of prime numbers, possesses this cardinal number, and any two different series of this set give rise to different groups g.

We can sum up the preceding as follows:

THEOREM 5. *The geometric type of order* ζ *allows of an infinite number of groups consisting of a geometric type of order* ζ *of continuous one-one transformations and being uniformly approximated by a fundamental series of groups consisting each of a finite number of continuous one-one transformations.*

If in particular we consider those groups g for which each g_n is chosen in the way described at the commencement of this § as the direct product of g_{n-1} and a group γ_n, we can formulate in particular:

THEOREM 6. *The geometric of order* ζ *allows of an infinite number of groups consisting of a geometric type of order* ζ *of continuous one-one transformations and being uniformly convergent direct products each of a fundamental series of finite groups of continuous one-one transformations.*

§ 5.

The sham-addition in the geometric type of order ζ.

Let us choose the factor groups indicated in theorem 6 as simply as possible, namely g_1 as the group of cyclic displacements corresponding to a certain cyclic arrangement of the first indices, and likewise each γ_n as the group of cyclic displacements corresponding to a certain cyclic arrangement of the n^{th} indices; g is then commutative, and transitive in such a way that a transformation of g is determined uniformly by the position which it gives to one of the elements of μ.

Let us further choose an arbitrary piece of μ as *piece zero*. Let us represent this piece by S_0, and the transformation, which transfers S_0 into S_α and is thereby determined, by "$\mp S_\alpha$". That the

piece S_β is transferred by this transformation into S_γ, we shall express by the formula

$$S_\beta \dot{+} S_\alpha = S_\gamma,$$

which operation is associative and commutative.

Let us finally choose, in order to make the resemblance to ordinary ciphering as complete as possible, all m_n's equal to 10, let us take for each system of n^{th} indices the digits 0, 1, 2, 3, 4, 5, 6, 7, 8, 9 in this order, and let us give to the piece zero only indices 0.

The different pieces of μ we can then represent biuniformly by the different infinite decimal fractions lying between 0 and 1, in such a way, however, that *finite* decimal fractions do not appear and that $\cdot 3\dot{0}$ is not equal to $\cdot 2\dot{9}$, whilst each group γ_n consists of the different ways in which one can add the same number to all n^{th} decimals, modulo 10.

Now according to the above we understand by $\cdot 5473 \ldots \dot{+} \cdot 9566 \ldots$ the decimal fraction, into which $\cdot 5473 \ldots$ is transferred by the transformation which transfers $\cdot\dot{0}$ into $\cdot 9566 \ldots$, or, what comes to the same, the decimal fraction, into which $\cdot 9566 \ldots$ is transferred by the transformation which transfers $\cdot\dot{0}$ into $\cdot 5473 \ldots$

We shall call the operation furnishing this result, on the ground of its associativity and commutativity, the "*sham-addition*" of $\cdot 9566 \ldots$ to $\cdot 5473 \ldots$; it takes place just as ordinary addition, with this difference that in each decimal position the surplus beyond 10 is neglected, thus that different decimal positions do not influence each other. So we have:

$$\cdot 5473 \ldots \dot{+} \cdot 9566 \ldots = \cdot 4939 \ldots$$

Let us understand analogously by $\cdot 5473 \ldots \dot{-} \cdot 9566 \ldots$ the decimal fraction, into which $\cdot 5473 \ldots$ is transferred by the transformation which transfers $\cdot 9566 \ldots$ into $\cdot\dot{0}$, and let us call the operation furnishing this decimal fraction the "*sham-subtraction*" of $\cdot 9566 \ldots$ from $\cdot 5473 \ldots$; then this sham-subtraction is performed in the same way as ordinary subtraction with this difference, that "borrowing" does not take place at the cost of the preceding decimal positions, so that here again different decimal positions do not influence each other. So we have:

$$\cdot 5473 \ldots \dot{-} \cdot 9566 \ldots = \cdot 6917 \ldots$$

By operating only with a finite number, great enough, of consecutive figures directly behind the decimal sign, sham-addition and sham-subtraction furnish in the type of order ζ a result agreeing with the exact one up to any desired degree of accuracy. In this too they behave like ordinary addition and subtraction of real numbers.

Microbiology. — *"Emulsion laevulan, the product of the action of viscosaccharase on cane sugar"*. By Prof. M. W. BEIJERINCK.

In the proceedings of the Academy of 9 February 1910 an enzyme was described which produces from cane sugar and raffinose a viscous matter incapable of diffusion. My further investigations, made conjointly with Mr. D. C. J. MINKMAN, proved that this substance is closely related to the laevulan of LIPPMANN [1]) but not identic with it.

Our emulsion laevulan originates in watery nutrient solutions in quite the same way as in the agarplates, so that these solutions change into a milkwhite emulsion; the liquid between the suspending laevulan droplets opalises very strongly. In hot water the substance is fairly soluble and the specific rotation of the polarised light, which, on account of the opalisation can only approximately be determined, is about

$$\alpha_D = -80°$$

whilst LIPPMANN gives for his laevulan

$$\alpha_D = -221°.$$

On account of this considerable difference in its rotating power, a new name, e.g. "sinistran", might seem desirable. But the word laevulan having a collective meaning to which also the more and the less soluble forms of our substance may be brought, we shall here use the general denomination, the more so as it is sure that the laevulan of the literature, like ours, consists of the cell-wall substance of bacteria.

Besides by LIPPMANN the formation of laevulan by bacteria has also been observed by MAASSEN [2]), who does not, however, describe the appearance of the emulsion, so that in this case, too, a modification of our emulsion laevulan seems to be produced. The here concerned microbe is a sporulating fermentation organism, called by MAASSEN *Semiclostridium commune*, but not yet found by us.

Preparation and properties of emulsion laevulan.

We were first of opinion that emulsion laevulan might be best prepared by using *Bacillus emulsionis*, for we had stated that this species does not decompose the once formed laevulan, whilst *B. megatherium* and *B. mesentericus*, which likewise produce emulsion

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[1]) Chemie der Zuckerarten 3te Aufl. 1904. Pag. 906, 1312.

[2]) Arbeiten aus dem Kaiserl. Gesundheitsamte. Biol. Abt. Bd. 5, p. 2, 1905.

laevulan, attack this substance and use it as food as soon as the cane sugar fails. We have, however, found that with some precaution it is much easier, especially with *B. mesentericus*, to produce large quantities of laevulan, than with *B. emulsionis*; this reposes on the circumstance that the former species, particularly at high temperatures, about 40°, possesses a very strong vegetative power, whilst the latter always grows slowly and has a relatively low temperature optimum, below or near 30° C.

Hence we used for the preparation of laevulan the common hay bacterium, which is the form of *B. mesentericus* obtained by accumulation methods, such as the method of potato slices and that of malt solutions. But this form is so common in our surroundings and so well adapted to the life in cane sugar solutions of for the rest different composition, that these, after pasteurisation or short boiling and when kept warm, of themselves produce laevulan by the development of the spontaneous spores of the hay bacillus. Such solutions then turn milky and slimy by the formation of the microscopic laevulan emulsion.

For the experiments were used large ERLENMEYER-flasks with 500 cm^3 of a medium of the composition: tapwater, 20 % canesugar, 0.05 % KNO_3, and 0.05 % K_2HPO_4, cultivated at ± 27° C.

This liquid inoculated with *B. mesentericus* very soon obtains the said milky appearance. The same emulsion which to the colonies of *B. mesentericus* and *B. emulsionis* on cane sugar agarplates gives so peculiar a character, is now in large quantity produced in the culture liquid, saturated besides with laevulan in true solution, which causes the strong and characteristic opalisation, not known to us to such a degree in any other substance. Besides, at the bottom of the flasks a thick transparent slime layer is slowly formed, which also proved to consist of laevulan, wherein, however, the bacterial bodies themselves are accumulated, whilst the liquid above it is poor in bacteria but abounds in viscosaccharase and laevulan emulsion. The acid formation in this solution is slight but not absent.

The laevulan may be precipitated with alcohol for which 50 % in the solution is sufficient. Only at a much greater alcohol concentration other substances of the liquid also precipitate. By dissolving in boiling water and again precipitating the further purification is easy. After drying and pulverising a snowwhite nearly tasteless powder results.

From a flask as the above which at first contained 100 G. of cane sugar, 8 G. of pure dry laevulan were obtained after 7 days cultivation, there still being in the liquid 20 G. of invert- and 70 G. of cane sugar; the slime at the bottom not being collected.

From another flask quite alike to the preceding, which also contained 100 G. of cane sugar, were gained 15 G. of laevulan after 17 days, 45 G. of cane sugar and 35 G. of invert sugar still being present.

The slime adhering to the bottom, consisting of *B. mesentericus* with thick cell-walls of laevulan, was used for a new culture for which a solution of 2 % of cane sugar, 0.05 % KNO_3 and 0.05 % K_2HPO_4 was used. After 18 days were obtained 2.25 G. from the 10 G. of original cane sugar, accordingly 22,5 % of laevulan was earned.

Pure laevulan is somewhat soluble in cold water, much better in boiling; all solutions opalise very strongly. It does not reduce FEHLING's coppersolution; only after prolonged boiling a feeble reduction is observed. It is incapable of alcoholic and lactic acid fermentation, but by butyric acid ferments, in absence of air, it gets into as strong a fermentation as cane sugar, whereby hydrogen, carbonic and volatile acid result.

A number of bacteria can feed on it when growing with access of air. *Azotobacter chroococcum* can use it under fixation of free nitrogen and formation of some acid.

By a treatment with acids, especially when warm, it changes readily into laevulose and so becomes fit for alcoholic and lactic-acid fermentation. After the inversion, by heating with resorcine and strong hydrochloric acid, the red colour appears, characteristic of laevulose, whilst with orcine and hydrochloric acid the violet colour, indicating pentose, is completely absent. When distillated and treated with sulphuric acid no perceptible quantity of furfurol can be detected.

As said, the specific rotation, which cannot be exactly determined on account of the strong opalisation is

$$\alpha_D = -80°,$$

and after hydrolysis

$$\alpha_D = -70°.$$

After prolonged heating with acid in the autoclave at 120° the rotation lowered even to

$$\alpha_D = -64°.$$

That of pure laevulose is

$$\alpha_D = -92°.$$

There is some probability that this diminution is due to destruction of part of the laevulose.

As we had found that the slime at the bottom of the flask is less soluble than that obtained by alcohol from the emulsionated liquid above it, we prepared laevulan from this slime also by separate experiments, for we supposed that dextran might occur therein, which is much less soluble in water than laevulan. However, it was found that the laevulan obtained in this way gives no other rotation after inversion than the emulsion laevulan, from which it does not differ. Hence it is sure that hay bacteria produce no dextran at all, but that their cell-wall consists of various modifications of laevulan of different solubility.

Not only in media of the above composition *B. mesentericus* produces laevulan, this happens quite as easily in a yeast decoction with 2 to 20 % of cane sugar, addition of chalk proving favourable. The temperature of cultivation may also vary and even rise to 40° C., but then care should be taken that the laevulan itself be not attacked by the producer.

From the preceding it may be concluded that the large lumps of slime so easily formed on cane sugar agar-plates by *B. mesentericus* and the other emulsionating species consist as well of laevulan as the emulsion which occurs round the colonies of this species in the agar. Hence, it can neither be doubted that the slime of these colonies, which does not diffuse in the agar, is produced by viscosaccharase from cane sugar, and that this enzyme only partly gets out of the bacterial body proper, the cell-wall included. Evidently in the cell-wall itself the enzyme forms new laevulan by converting the cane sugar, with which both cell-wall and agar-plate are imbibed.

The production of cell-wall substance in consequence of the action of an enzyme, which in my former communication was called probable, must now, as regards laevulan, be considered as proved.

Dextran and the dextran bacteria, which we have likewise studied, shall later be treated more thoroughly. For the moment it may be observed that by this substance the polarised light is strongly rotated to the right; we found

$$\alpha_D = + 132°,$$

whilst in the literature by various authors is given for dextran

$$\alpha_D = + 199° \text{ to } 230°.$$

Quite like laevulan it results exclusively from cane sugar. So laevulan as well as dextran are produced by microbes, neither from laevulose, glucose, or any other sugar, but solely from cane sugar and raffinose. The slimy cell-wall substances formed by other microbes from glucose, laevulose and maltose, are of a different nature.

Physics. — *"Researches on the magnetization of liquid and solid oxygen."* By H. KAMERLINGH ONNES and ALBERT PERRIER. Communication N°. 116 from the Physical Laboratory, Leiden.

§ 1. *Introduction.* It is scarcely necessary to remark that the investigation of the magnetic properties of oxygen at low temperatures has long occupied a position on the programme of the cryogenic laboratory, or that this has been considered one of the most important items on the programme since the investigation of both liquid and solid oxygen has been made possible by the perfecting of the methods [1]) of obtaining detailed series of measurements at constant temperatures in the region of liquid hydrogen. Indeed, while the strongly magnetic properties of oxygen of themselves select it from all other substances as especially suitable for the study of para-magnetism, we can in the meantime for no other substance obtain the *magnetic equation of state* [2]), which gives a representation of the magnetic properties of a substance in the three states of aggregation at as many successive temperatures and pressures as possible.

The investigation of oxygen at very low temperatures and at pressures that can easily be realised was expected to give at once results of much importance.

CURIE [3]) found for gaseous oxygen between 20° C. and 450° C. that the *specific susceptibility* (magnetization per gram for $H = 1$) was inversely proportional to the absolute temperature, and FLEMING and DEWAR [4]) concluded from their latest measurement of the susceptibility of liquid oxygen at its boiling point that CURIE's law was obeyed down to —183° C.

Does the specific susceptibility continue to increase so strongly at low temperatures or does it approach a limiting value? Is oxygen in the solid state ferro-magnetic? Does the magnetization finally at extremely low temperatures perhaps begin to decrease and disappear completely at the absolute zero? [5])

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[1]) H. KAMERLINGH ONNES, These Proc. Sept. 1906, Comm. from the Leyden labor. no. 94*f* (1906).

[2]) H. KAMERLINGH ONNES, Commun. from the Leyden labor. Suppl. no. 9 p. 28.

[3]) P. CURIE. Ann. chim. phys. (7) 5 (1895) p. 289.

[4]) FLEMING and DEWAR Proc. Royal Soc. London 63, p. 311, 1898.

[5]) It has since appeared that the magnetization of ferro-magnetic substances does not yet give any justification when the temperature is lowered to the melting point of hydrogen for the assumption that the electrons whose motion causes magnetization are frozen fast to the atoms and that therefore this disappearance at the absolute zero may be expected. (P. WEISS and H. KAMERLINGH ONNES, These Proc. Jan./Febr. 1910, Comm. from the Leyden Labor no. 114 p. 9).

These are questions which, considering the possibility of obtaining important contributions to the knowledge of the influence of density upon susceptibility by high pressures in the region where the gaseous state of aggregation changes continuously into the liquid make it a very attractive work to realise, even from a purely experimental point of view, the representation to which we have just referred.

The work was commenced though only when Prof. WEISS extended his magnetical researches to very low temperatures and the measurements on the magnetization of ferro-magnetic and cognate substances at very low temperatures, which were communicated to the February Meeting [1]), were undertaken. With that investigation which was carried out at the same time, the present one is very closely related, and for part of them we made use of the same appliances. In our present investigation we have also in various ways made use of Prof. WEISS's method [2]) of determining the magnetization by means of the maximum couple exerted by a magnetic field of variable direction upon an ellipsoid of the experimental substance, a method which had been entirely successful in the other research. We must also express the great advantage we derived from the continued support given us by Prof. WEISS, and we take this opportunity of gratefully acknowledging our indebtedness to him.

The change with temperature of the specific susceptibility of oxygen, the investigation of which was our first object, is of particular importance seeing that CURIE's law follows from LANGEVIN's kinetic theory of magnetism [3]). It was soon apparent to us that this law was not valid for oxygen, as was thought, down to — 183° C., but that it would have to be replaced by another. According to the important paper of DU BOIS and HONDA communicated to the January Meeting — our experiments had already been completed at that time — various elements were found for which CURIE's law did not hold at temperatures above 0° C. This at once increases the importance of the further investigation of oxygen, for which over a definite region of temperature CURIE's law is valid, while over another region it obeys a second law, viz.: *that of inverse proportionality to the square root of the absolute temperature.* The results concerning this law and also concerning the probability of a *sudden change in the value of the specific susceptibility on solidification* will be discussed in § 5.

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1) P. WEISS and H. KAMERLINGH ONNES. These Proc. Jan./Febr. 1910. Comm. fr. the Leyden labor. no. 114 (1910).

2) P. WEISS. Journ. de phys. 4e série t. VI, p. 661; 1907.

3) LANGEVIN. Ann. chim. phys. (8) 5, p. 70; 1905.

We have been occupied with another question besides the change of specific susceptibility with temperature, which was suggested both by the experimental results obtained by FLEMING and DEWAR and by the theories of LANGEVIN and WEISS.

In the experiments of the first-named there appears sufficient evidence for the conclusion that there is a decided diminution of the susceptibility as the strength of the field increases (the diminution is of the order of 10% in a field of 2500 gauss). Now, according to the theory of LANGEVIN para-magnetic substances must, it is true, exhibit this phenomenon, but calculation from his formulae limits the magnitude of this change to less than 0.1% in the case of liquid oxygen at its boiling point. Should a higher value than this be obtained, then one would be led to assume the existence of a WEISS molecular field [1]). We arranged our experiments so that the liquid and the solid oxygen could be subjected to a field of 16000 gauss, a field very much stronger (about six times) than that used by FLEMING and DEWAR, so that we might expect the phenomenon which appeared in the course of their experiments to be exhibited to a much greater degree in ours even at the same temperatures as were used by them. If what was observed by FLEMING and DEWAR could really be ascribed to the beginning of saturation then the theory would further lead us to expect that as the temperature sank the change would strongly increase (becoming infinite at $T = 0$), and that in our experiments with liquid hydrogen it would become very striking. We have, however, observed only small deviations, which we shall discuss further in § 5.

As regards the experimental methods employed by us in our investigation, two completely different schemes were adopted: on the one hand was measured the magnetic attraction exerted upon a column of the liquid, and on the other the maximum couple exerted by a homogeneous field upon an ellipsoid. The second method is more especially suitable for comparative measurements and can also be used for frozen oxygen; the first can be used only for the liquid phase, but on the other hand it makes very trustworthy absolute measurements possible; we have therefore adopted it as the basis of our other measurements. In the carrying-out of each method further precautions are still desirable, so that while we are busy pushing on the investigation, we propose at the same time to repeat it in part in order to increase the accuracy of the results obtained by taking such further precautions as have appeared possible in the course of the work.

[1]) WEISS, L'hyp. du champ moléc. loc. cit.

Liquid oxygen I.

§ 2. *Method of the magnetic rise.* As mentioned above, we have rendered the method of the magnetic rise employed by QUINCKE, DU BOIS and other observers suitable for use at low temperatures.

One limb of a vertical O-shaped tube, the upper portion of which contains the gaseous, and the lower the liquid phase of the experimental liquefied gas was placed between the poles of a magnet whose field was horizontal.

Let H be the field, (H' the field in the other limb is supposed to be so small that $(H'/H)^2$ is negligible), g the acceleration due to gravity, z the difference in height of the levels of the liquid under the influence of H, ϱ and ϱ_0 the densities of the liquid and of the gaseous phases respectively, K and K_0 their respective volume susceptibilities, then

$$(K-K_0)\,H^2 = 2z\,(\varrho-\varrho_0)\,g \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

or, by introducing the absolute specific susceptibility χ.

$$(\chi\varrho-\chi_0\varrho_0)\,H^2 = 2z\,(\varrho-\varrho_0)\,g.$$

If $\chi = \chi_0$ then the equation becomes simply

$$\chi = \frac{2zg}{H^2}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

which is the formula we have used for our calculations. [1])

So there are striking advantages offered by this method particularly for an absolute measurement, on account of its applicability to the case of a liquid in equilibrium with its own vapour. There are only two magnitudes to be determined, the distance z, which can be measured very accurately with a cathetometer, and the field H; nor have we to know the density of the liquid in order to be able to find the specific susceptibility.

Magnetic rise apparatus. It is a very easy matter to cause an ordinary liquid to ascend under the influence of magnetic attraction, but the experiment is attended by serious difficulties when one has to deal with a liquefied gas. Boiling must be completely avoided, and care must be taken that the vaporization is unnoticeable. The first precaution is necessary because the motion of the liquid or of its surface would render adjustment quite impossible, and the second

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[1]) In § 5 we shall give the reasons why we think that $\chi = \chi_0$, and should it be possible that this is not the case there is still the greatest probability that $\chi_0 < 1.5\,\chi$; in the most unfavourable case at the boiling-point the correction remains below 0.002 in value, while at lower temperatures it is quite negligible on account of the small value of χ_0.

is necessary that the total quantity of liquid may not appreciably alter during the measurement of one rise. Moreover magnetic action itself increases the difficulties; it is easy to see that it can occasion the formation of gas-bubbles which divide the column of liquid into two parts, so that the one portion remains suspended between the poles, while the other falls back again. In that case measurement of the ascent is out of the question.

Starting from the thermodynamic potential it appears that in every case the relation

$$2g\,y\,\varrho > K(H^2 - H_y^2)$$

must hold, where H is the field at the surface of the liquid, and H_y the field at a distance y below the surface of the liquid. These conditions shew that there is a limit to the intensity of the fields in which measurements may be made, for they necessitate a range of extended fields (in this case in a vertical direction). Conical pole-pieces are thus as a matter of fact barred.

After several preliminary experiments an apparatus was constructed, the most important part of which consisted of two concentric double-walled vacuum tubes, with which we already succeeded in obtaining rather successful measurements. The walls of the double vacuum tube were not silvered, so that we were able to watch how the liquid behaved during the experiments. From the experience thus acquired the improved apparatus which we shall now proceed to describe was designed and constructed.

It will be seen that the construction of the apparatus lays a very heavy tax upon the art of the glass-blower [1]). As before, the chief part consisted of two independent U-shaped vacuum tubes, the one fitting inside the other. The double walls of each tube are completely silvered on the vacuum side, except in the case of the inner tube, where the distance which the liquid ascends is left free, and in the outer where a sufficient length is left unsilvered to leave a strip of a few millimeters breadth through which the level of the liquid can be read. One of the tubes completely surrounds that portion of the other which contains liquid; this we call the protecting tube. The narrowest portion M_4 (fig. 2) is placed between the poles of the electro-magnet. The narrow limb of the inner tube must of course be perfectly cylindrical. The other limb is enlarged and serves as a reservoir. In order to be able to apply equation (2) all care was taken that the temperature of the liquid and vapour up to a height

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1) The double vacuum tube was prepared by Mr. KESSELRING, Laboratory glass-blower, and the remainder by Mr. FLIM, technical assistant at the Laboratory.

somewhat greater than that reached by the column of liquid was everywhere the same both in the wide and in the narrow tube; and further care was taken that where the temperature of the vapour above the liquid in the upper parts of the apparatus changes to ordinary temperature it was as far as possible the same at the same height in the two limbs of the O-shaped space. With this end in view the liquid in the inner tube was, by means of the magnetic field, repeatedly moved up and down under constant vapour pressure, until we might assume that in this tube equilibrium was sufficiently well attained. To make this equilibrium possible the inner tube is surrounded with liquid at the same temperature as that which the liquid in it must attain. In the outer or protecting tube the liquid is kept constantly in motion by means of a stirrer consisting of a brass ring S_1 that can be moved up and down; it is possible to do this and still keep the space closed by utilising the flexible rubber tube S_2. The vaporization in the inner tube is thus very small (between 0.5 and 1 litre of gas measured under normal atmospheric pressure escapes per hour).

Notwithstanding all these precautions temperature differences must still be encountered. In the liquid, in which the convection currents maintaining heat-equilibrium can be followed by the small particles which they carry along with them, these temperature differences must have been very small. In the gas layer in the upper portion of the O-shaped space there must indeed have been considerable differences; but on account of the small density of the gas, these have but small influence upon the difference of level in the two limbs, and, moreover, that influence may be almost entirely neglected seeing that the observations are simply comparative measurements with and without the magnetic field. Now, care has been taken that the temperature over the distance that the liquid rises can vary but slightly, while in the upper portions of the tube practically the same state of affairs is maintained during both observations. We have therefore omitted the correction that should still have to be applied for possible temperature differences.

Comparing the positions of the liquid in the narrow cylindrical tube with and without the magnetic field also reduces the correction for capillarity to the insignificant differences in form of the menisci, and this correction, too, we have omitted.

The inner and the outer tubes are closed independently of each other by means of the German-silver caps P_1, P_4, Q_1, Q_4 (fig. 1); the junction is made air-tight by the rubber sleeves M_0, N_0, which at the same time unite the two tubes firmly together. Liquid oxygen

is introduced into the protecting tube through the small tube P_5, and into the inner tube through Q_5. The two tubes P_3 and Q_3 lead the vaporized oxygen through the valves P_3 and Q_3 (fig. 2) to two gasometers. Two manometers P_6 and Q_6, the latter of which is provided with an indicator Q_7 so that small vapour pressures may be read off accurately, serve at the same time as safety valves. It is not necessary that the oxygen in the protecting tube should be as pure as that in the inner tube; for the latter, with which the observations were made, very pure oxygen was used.

A double sliding movement R allowed an easy adjustment of the apparatus each time, so that the meniscus in the measuring tube just reached the desired point in the field between the poles, usually in the axis of the pole pieces.

Course of a series of measurements. The field is brought to the desired strength and by means of R the meniscus is made to rise to the desired point, which is read off on a small scale. Then the meniscus is moved up and down several times while care is taken that the field slowly increases. In this way the temperature is made everywhere the same and the walls of the tube are wetted. While the field has the desired value the position of the meniscus is read off; then a reading is made while the field is off; after the meniscus has been three times allowed to rise somewhat higher than the desired position, another reading is made while the field is on; once more a reading is made with the field off and so on several times. In this manner the error arising from vaporization of the liquid during the adjustment of the cathetometer is eliminated [1]). It is not essential to know the position of the level in the other limb of the tube; so as to be able to take account of this, we ascertained the ratio of the cross-sections of the two limbs of the tube.

We have further made sure that the residual magnetism exerted no appreciable influence upon the position of the meniscus after the current was cut off. For this purpose a feeble current was sent through the coils in the direction opposite to that which had just been broken. Had the residual field exerted any appreciable influence we should have seen first a further sinking of the level, and then a rise as the current was slowly increased. This has not been observed.

We used the same electro-magnet as was used for the cryogenic investigation of the ferro-magnetic metals [2]), to which we must refer

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1) To control the position of the meniscus without the magnetic field, we measured the quantity of gas vaporized (cf. preceding page).

2) P. Weiss and H. Kamerlingh Onnes, l. c.

for details regarding its construction. It was only necessary to replace the conical pole-pieces by cylinders with flat ends. Their distance apart was micrometrically adjusted to 25 mm. and controlled with an accurate callipers. We may here remark that between the measurement of the ascent and that of the field, the pole-pieces remained clamped tight to the cores, so the adjustment of the distance could give rise to no error.

Since in the subsequent calculation the strength of the field is involved to the second power, and since we are concerned with an absolute measurement, we endeavoured to make our measurement of the field strength as trustworthy as possible with our present appliances. With this end in view we measured the strength of an arbitrarily chosen standard field by two different processes, and we compared the strengths of the fields used in our experiments with this standard by successively withdrawing the same coil attached to a ballistic galvanometer from the standard field and from the various fields which we desired to measure.

The standard field was set up with the same flat pole-pieces at a distance of 9 mm. apart, and with a current of 5 amp. All precautions were taken to ensure the demagnetization of the magnetic cycle beforehand. This field was first measured by means of COTTON's magnetic balance[1]). As is well known this method consists of equilibrating weights of a total mass m against the ponderomotive power of the field H on a straight portion of length l of a conductor through which a current flows of intensity i; then we get

$$H = \frac{m\,g}{i\,l}\,.\,10.$$

For the degree of accuracy, however, which we wish to reach, several corrections must be taken into account. In the first place the various parts of the balance were accurately calibrated. The length l of the current element was determined micrometrically and on the dividing engine, and so also was the distance between the arcs of the balance which distance ought to be the same throughout seeing that the arcs must be accurately concentric. The very small deviations from this were allowed for by means of a ballistic investigation of the topography of the field. The balance arms of the weights and of the current element were measured with the cathetometer. The topographical study of the field also gave us the

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[1]) For this method of measuring the field and for the magnetic balance see: P. WEISS and A. COTTON, Le phénomène de ZEEMAN pour les trois raies bleues du zinc, Bull. Séances Soc. franç. de phys. 1907, p. 140, also J. de phys. 1907.

correction necessary for the force exerted upon the second straight element of the balance (i. e. that outside the pole-gap). The sum total of these positive and negative corrections came to some units per thousand.

The greatest care had to be devoted to the absolute value of i, which was measured by means of an accurate ammeter by SIEMENS and HALSKE. This was calibrated in absolute ampères by comparing on the potentiometer the potential difference between the terminals of an international ohm (or for the stronger currents of 0,1 Ω) with the electromotive force of a WESTON cadmium cell. For the requisite accuracy of the measurements the influence of neighbouring instruments or currents upon the ammeter, or of its position in the earth's field were by no means negligible; we got rid of almost all these irregularities by a suitable adjustment of the distances and of the positions of the rheostats, and we eliminated further possible remaining errors by so connecting *all the conductors* that the currents *in all* except the ammeters could be reversed *at the same time*. Finally we always used the ammeters in the same position with respect to the earth's field as that in which they had been calibrated.

When all calculations and corrections had been completed it was found that the strength of the standard field was 9857 gauss according to this method.

The *second method* by which the value of the same field was found consisted of the sudden withdrawal from between the poles of the magnet of a coil of wire of which the area encircled by the current was known. The change thus caused in the number of induction lines embraced by the coil was compared by means of a ballistic galvanometer with the number of induction lines embraced by a solenoid the dimensions of which were accurately known.

The coil consisted of 19 turns of silk-insulated wire, 0.25 mm. thick, wound round a cylinder of ebonite, 20 mm. in diameter. The dimensions were obtained by various measurements with the micrometer screw and the dividing engine, and were repeatedly controlled. At the same time a control coil was constructed by winding bare copper wire in a helical groove cut in the curved surface of a cylinder of ebonite; the area encircled by the current was then measured for this control coil by the same methods and with the same precautions as were adopted in the case of the first. The ratio of the two was in agreement with the ratio of the deflections of the ballistic galvanometer which were obtained by connecting the two coils in series with the galvanometer and then withdrawing them successively from an unchanged magnetic field. We may further

say that we had previously verified the absence of magnetic properties from the ebonite by means of an apparatus after CURIE in which we utilised the attraction in a non-uniform field.

For the measurement of the field there were placed in circuit with the galvanometer the coil on the ebonite cylinder, a manganin resistance to regulate the sensitivity, a secondary coil of 500 turns fitting round the standard solenoid, and finally, an electromagnetic arrangement which could be used as a damper if desired. We also allowed for the very small deviations from the law of proportionality between the deflections of the galvanometer and the quantities of electricity, which had been determined for the galvanometer (one of the DEPREZ-D'ARSONVAL type) by a previous investigation. The solenoid was constructed with the greatest accuracy by winding bare copper wire on a core of white marble [1]).

The standardisation of the galvanometer was made by reversing the current in the solenoid; the observations made by withdrawing the coil from the field always took place between two standardisations of the galvanometer; there was, however, no change in the galvanometer constant to be observed. The corrections and precautions necessary in obtaining the strengths of the current are the same as in the case of the balance, and have already been described. The final result of this ballistic method is

9845 gauss.

The relative difference between this and the value given by COTTON's balance is therefore 0.0012; and this can be neglected especially when one remembers that almost every one of the numerous measurements necessitated by the one method as much as by the other, beginning with the adjustment of the field by means of the ammeter, is accurate only to 0.0005. It may be useful to comment here upon a particular point that increases the difficulty of obtaining this agreement and therefore enables us to rely more upon the correctness of the numbers which we have obtained. The equation for COTTON's balance involves the strength of the current in the *denominator*, while this magnitude in calculating according to the ballistic method occurs in the *numerator*; a systematic error there-

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1) For the dimensions and the description of the solenoid and galvanometer see: P. WEISS, Mesure de l'intensité d'aimantation à saturation en valeur absolue, Arch. Sc. phys. et nat. February 1910, J. de phys. May 1910.

fore in the *absolute* number of ampères would, of necessity, occasion a relative difference *twice as great* between the values of the field obtained by the two methods (the same ammeter was used with the balance and with the solenoid).

We have given the ballistic method a somewhat greater weight than the other on account of the smaller number of corrections it involved, and thus we have finally taken as the value of the standard field

9850 gauss.

Once this standard field was definitely fixed all other measurements could be rapidly made by the ballistic method described above.

For the conical pole-pieces which are employed in experiments according to the maximum couple method, and which give much more powerful but much less uniform fields, we used a coil of 7 to 8 mm. diameter accurately centred on the axis of the pole-pieces. In this case direct comparison with the standard field just mentioned was not possible since the flat polepieces had to be screwed off to make room for the conical poles. To meet this case the area of the small coil encircled by the current was determined once and for all by withdrawing it from the standard field *before* the flat pole-pieces were removed, and comparing the change thus brought about in the number of the induction lines with those of the solenoid by means of the ballistic galvanometer.

All the measurements that we have given up to the present refer to the field in the centre of the space between the poles. For the few exceptional values of the field, and, consequently, of the ascent of the liquid oxygen for which it was necessary to cause it to rise pretty far above the axis of the pole-pieces, the field was determined at those points by simple ballistic comparison with the fields on the axis, and we made use of the cathetometer to adjust the position of the small coil.

Results of observations and calculations.

Series of observations with the apparatus with unsilvered walls.

TABLE Ia. $t = -183°.0$ C. [1])

Position of meniscus.	Obs. rise z' in cm.	diff. in height with reservoir z in cm.	H in gauss.	$\frac{z}{H^2} . 10^7$
Level of axis	1.032	1.061	2980	1.194
"	1.646	1.690	3727	1.216
"	1.656	1.701	3727	1.224
axis + 2.44 cm.	3.024	3.110	5182	1.158
axis	3.198	3.289	5205	1.214
"	4.16	4.278	5848	1.251
axis + 2.44	4.90	5.050	6570	1.168
axis	5.124	5.270	6600	1.210
axis + 2.44	7.87	8.094	8075	1.242
axis + 2.44	9.20	9.462	9043	1.158*

The difference in height z was obtained from the observed ascent from $z = z'(1 + 0.0285)$. The observation * was very difficult and is little reliable.

The deviation in the case of the observation in a field of 5182 gauss is probably due to a mistake of 1 in the number of whole millimeters which were read off, but of this we are not certain.

Deviations from proportionality with H^2 are considerable but by no means systematic. If we take the mean of all the measurements with the exception of the last in which special difficulties were encountered we reach the value

$$\frac{z}{H^2} = 1.209 . 10^{-7},$$

and for the specific susceptibility with $g = 981.3$ for Leiden

$$\chi_{90°.1\,\mathrm{K.}} = 237.3 . 10^{-6}.$$

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[1]) The boiling point of oxygen according to H. KAMERLINGH ONNES and C. BRAAK These Proc. Oct '08. Comm. fr. th. Leyden labor. N°. 107a § 6.

TABLE Ib. $t = -201°.75$ C.

Position of meniscus.	Obs. rise z' in cm.	diff. in height with reservoir z in cm.	H in gauss.	$\frac{z}{H^2} \cdot 10^7$
Level of axis	1.192	1.222	2980	1.376
"	1.893	1 944	3727	1.399
"	1.881	1.935	3727	1.393
"	3.643	3.747	5205	1.383
"	4.623	4.752	5848	1.389
"	5.91	6.078	6600	1.395
axis + 2.5 cm.	7.376	7.586	7421	1.378
axis + 2.5 cm.	8.715	8.963	8069	1.372

$$\text{mean} \quad \frac{z}{H^2} = 1{,}386 \cdot 10^{-7}$$

whence it follows that $\chi_{71°.35\ K.} = 272{,}0 \cdot 10^{-6}$.

Finally, at $-209°{,}2$ C. a single observation was made. The rise was 6.115 cm. in a field of 6600 gauss, which with the correction for the sinking in the reservoir gives

$$\frac{z}{H^2} = 1.444 \cdot 10^{-7} \text{ and } \chi_{63°.9\ K.} = 283.4 \cdot 10^{-6}.$$

We shall now give the series of observations made with the silver-walled apparatus which we have already described.

TABLE IIa. $t = -183°.0$ C.

Position of meniscus.	Obs. rise z in cm.	diff. in height with reservoir z in cm.	H in gauss.	$\frac{z}{H^2} \cdot 10^7$
axis	1.060	1.090	2980	1.227
"	1.669	1.716	3727	1.235
"	3.169	3.258	5183	1.213
"	3.220	3.310	5198	1.225
"	4.035	4.148	5807	1 230
"	4.093	4.208	5848	1.230
"	4.101	4.216	5848	1.233
"	5.119	5.262	6578	1.216
axis + 2.5 cm.	7.750	7.967	8075	1.224
"	8.950	9.201	8659	1 227
axis + 3.5 cm.	9 226	9.484	8808	1.222
"	9 266	9.525	8808	1.228

For this apparatus $z = z'(1 + 0{,}0280)$.

The mean value of $\frac{z}{H^2}$ is $1{,}226.10^{-7}$, whence it follows that

$$\chi_{90°.1\ K.} = 240.6.10^{-6}.$$

TABLE IIb. $t = -201°.75$

Position of meniscus	Obs. rise z' in cm.	diff. in height with reservoir z in cm.	H in gauss	$\frac{z}{H^2} \cdot 10^7$
axis	1.195	1.228	2980	1.383
"	1.879	1.932	3727	1.391
"	3.625	3.726	5205	1.375
"	4.567	4.695	5848	1.373
axis + 3.5 cm.	5 461	5.614	6399	1.371
axis + 2.5 cm.	5 832	5.995	6567	1.390
axis	5.852	6 016	6600	1.381
axis + 3.5 cm.	6.463	6.644	6986	1.365
axis + 2.5 cm.	6.899	7.092	7169	1.380
axis + 3.5 cm.	8.207	8.437	7863	1.365
axis + 2.5 cm.	8.654	8.892	8069	1.366
axis + 3.5 cm.	8.988	9.240	8212	1.370
axis	8.913	9.162	8212	1.358

Mean of all observations is 1.375 whence it follows that

$$\chi_{71°.35\ K.} = 269.9 \cdot 10^{-6}.$$

TABLE IIc. $t = -208°.2$ C.

Position of meniscus	Obs. rise z' in cm.	diff. in height with reservoir z in cm.	H in gauss	$\frac{z}{H^2} \cdot 10^7$
axis	1.277	1.313	2980	1.478
"	1.996	2.052	3727	1.477
"	3.813	3.920	5205	1.447
"	4.841	4.977	5848	1.461
axis + 2.5 cm.	6.012	6.180	6567	1.433
axis	6.094	6.264	6600	1.438
"	6.113	6.284	6600	1.443
axis + 2.5 cm.	7.146	7.346	7169	1.429
axis + 3.5 cm.	8.579	8.819	7863	1.426

Mean of all observations 1.448, whence it follows that

$$\chi_{61°.9\,K.} = 284.2 \,.\, 10^{-6}.$$

Finally for finding the specific susceptibility the density of oxygen was found from the formula [1])

$$\varrho = 1.2489 - 0.00481\ (T - 68).$$

From table II we obtain

$$K_{90°.1\ K.} = 275{,}2 \,.\, 10^{-6}$$

$$K_{71°.35K.} = 332{,}8 \,.\, 10^{-6}$$

$$K_{61°.9\ K.} = 359{,}0 \,.\, 10^{-6}.$$

Table III gives $\chi\sqrt{T}$ for each of the temperatures and for each of the series.

Series with the first apparatus			Series with the improved apparatus		
T	$\chi . 10^6$	$\chi\sqrt{T} . 10^3$	T	$\chi . 10^6$	$\chi\sqrt{T} . 10^3$
90.1	237.3	2.25	90.1	240.6	2.283
71 35	272.0	2.29	71.35	269.9	2.279
63 9	283.4	2.26	64.9	284.2	2.289
mean		2.27			2.284

There is no systematic change to be noticed in the product $\chi\sqrt{T}$; the greatest deviation from the mean is 1% with the first apparatus, and only $^1/_4$ % with the second; moreover the deviations in the two series at corresponding temperatures are in opposite directions. Hence within the limits of accuracy of the observations the specific susceptibility can be represented by the formula

$$\chi = \frac{2284}{\sqrt{T}} \,.\, 10 - \epsilon.$$

In the comparative measurements which we shall describe in the sequel we shall find the same law, at least as far as its form is regarded. For the discussion of this point we refer to § 5.

The differences between the various values of the ratio $\frac{z}{H^2}$ are greater than we should be led to expect from the accuracy obtained

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[1]) BALY and DONNAN. J. Chem. Soc. 81 (1902) p. 907.

(0,05%) in the measurements with the cathetometer of the displacements of the level, and from the accuracy of the measurements of the field-strengths, of which a discussion is given above. It is certain that the cause of these deviations must arise from a source other than the measurement or these two data, though we cannot with certainty indicate what this may be.

We may in the meantime remark that, at least in the case of the first series, the unsteadiness of the apparatus in the vertical direction in the not quite homogeneous field, and the slight inconstancy of the temperature have certainly been contributory causes of these deviations, since the second apparatus which was improved exclusively in these directions gave much more regular results. This remark, however, does not seem to account sufficiently for certain appreciable changes that occurred without any noticeable corresponding irregularity in the pressure or in the convection current of the liquid, while there was also no noticeable change in the shape of the meniscus.

Liquid oxygen II.

§ 3. *Measurements by the method of the maximum couple exerted upon an ellipsoid.* Further comparative measurements for liquid oxygen at various temperatures were obtained by means of the method of the maximum couple exerted by a uniform field upon an ellipsoid. This method has already been described and discussed in connection with the research on ferro-magnetic substances [1]); it will be sufficient to discuss the modifications which were found to be necessary owing to the particular circumstances under which the method had to be applied to the present research.

In the first place on account of the small value of the susceptibility it was necessary to make the couple to be measured as large as possible; with this end in view we chose an *oblate* ellipsoid of revolution, instead of a prolate; its axis of revolution was placed horizontal in a field which could turn round a vertical axis.

The ratio that is taken between the axes is not a matter of indifference; for a given major axis the couple, which is proportional to $(N_1 - N_2)v$, is a maximum for a ratio of the major to the minor axis that is only slightly smaller than 3; we have therefore taken this value of the ratio for the construction of the ellipsoids.

We used the same electromagnet as served for the measurements made by WEISS and KAMERLINGH ONNES (loc. cit.). Two pairs of pole-

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[1]) P. WEISS, J. de phys. (4) 6 (1907) p. 655. P. WEISS and H. KAMERLINGH ONNES, Comm. N°. 114 These Proc. Jan./Febr 1910.

pieces were used; first the cylindrical pole-pieces with quite flat end surfaces that had been used for the measurement of the magnetic rise, and then truncated conical pole-pieces the end surfaces of which (slightly concave, see in this connection p. 818) were 4 cm. in diameter, and the side surfaces of which were connected by convex surfaces of revolution to the cylinders that formed the cores; these were 9 cm. in diameter. These pole-pieces were constructed to give the strongest possible field when the distance between the poles was taken to be 20 m.m. By this means a field of about 16000 gauss was obtained.

Our observations were made with an ellipsoid that was diamagnetic with respect to the surrounding medium — a solid silver ellipsoid immersed in a bath of liquid oxygen. The ellipsoid was turned by the "Société genevoise pour la construction d'instruments de physique" from a block of very pure MERCK silver. A preliminary experiment showed that it was very slightly diamagnetic with respect to air, and that this was quite negligible with respect to the liquid oxygen. The axes were measured microscopically on the dividing engine; this gave

major axis $= 1.0973$ cm. and axis of revolution $= 0.3654$ cm.

Furthermore, two intermediate ordinates parallel to the axis of revolution were measured on the dividing engine, and they were found to be 2% greater than the corresponding ordinates of a perfect ellipse with the same axes. This deviation from ellipsoidal shape was confirmed by a direct determination of the volume from the weight and the density, which gave

0.2329 c.c.

while calculation from the dimensions of the axes gave

0.2308 c.c.

In the calculations we made use of the value 0.2329.

The cryogenic apparatus, essentially the same as that used by WEISS and KAMERLINGH ONNES is shown in Pl. I fig. 3. Once more we see the *cover B*, the *adjusting tube f'*, and the *holder b'*. The cover with its various parts: the cap with the stuffing-box *D*, glass tube *C*, window with plane parallel glass plate C_1, the system *BG* for adjusting the whole apparatus, the tension rods B_4 for supporting the DEWAR tube, the helium thermometer θ, the little screens to protect the upper portions of the apparatus from cooling, etc. is just the same as before. The DEWAR tube is of the same shape, but the lower portion is of greater diameter. The only difference between the adjusting tube *f'* and that which was used in the other investigations is that the lower portion f_3' is of greater diameter.

The *holder* and the *torsion spring* are, on the other hand, completely altered. On account of the smallness of the couple to be measured all foreign magnetic actions had to be eliminated as carefully as possible. Preliminary experiments showed us that a metallic holder could not be used, not only on account of the traces of para- or ferro-magnetic impurities that are never absent from workable metals but also on account of the difficulty of keeping the surface sufficiently clean; this difficulty was encountered repeatedly in the silver ellipsoid that we used in our experiments, and it is probable that the constant contact of the hands with iron tools plays a part in causing it. Glass seemed to be by far the most suitable material both on account of the absence of inherent magnetization and of the fact that the surface on account of its smoothness can be kept quite clean. The holder which we finally adopted was made completely of glass: it consists of a tube b' 5 mm. in diameter that at b'_3 is drawn out to a narrow but thickwalled stem, 0.7 mm. in diameter. To this stem the silver ellipsoid was attached; for this purpose a hole of sufficient width to fit was bored along one of its greater diameters and the ellipsoid was then fixed at the desired height by means of a little wax that completely filled the narrow space between the glass and the metal. The tube was then pumped free from air and sealed off, so that the liquefaction of the air that it would otherwise contain would be prevented. The flat mirror for measuring the angle of torsion and the oil-damper were also attached to the holder.

The torsion springs. On account of the smallness of the couples to be measured (the constants of the springs were of the order of 1200 c.g.s. while those used for the investigation of the ferromagnetic substances were some tens of thousands) it was found more suitable to use a straight instead of a helical spring. We took a strip of phosphor bronze about 5.5 cm. long (l') and 0.2×0.01 sq. cm. in cross-section. The upper end was soldered to a spiral spring of three turns made from a much thicker strip than the other; the greatest dimension of this strip was horizontal so that in this way it fulfilled its purpose of being elastic to tension while taking no part in torsion; its presence is essential to prevent the breaking of the thin glass stem or of the platinum-iridium stretching wire that is soldered to the stem. This stretching wire is made from a platinum-iridium wire of 0.1 mm. diameter, which was rolled very thin so as to make its torsion constant extremely small without diminishing to any great degree its resistance to breakage. The stretching wire is fused at b'_4 to the lower end of the glass stem, and at its other extremity it carries a knob c' which is held fast in a ring f''_6.

The mounting of the apparatus took place with the same precautions regarding the centring of the whole, the tension of the springs, etc. and by a method similar to that which has been described in the research upon the ferro-magnetic metals.

The course of the observations is very simple once everything has been set up in position. First, those azimuths of the electromagnet are tentatively determined for which the couple in both directions is a maximum. It was sufficient to do these experiments two or three times with suitably chosen fields, since the azimuth changes but very little with the field, and for other values of the field one can without danger have recourse to interpolation. After that the series of observations took place in the following manner: Before making a measurement with any particular current this was reversed a certain number of times so as to obtain a well-defined field; we had not here to deal with a value of the saturation-magnetization, which changes but slowly with the field, but in our case the couple was proportional to the square of the field, so that inaccurate values of the field that might be obtained notwithstanding the fact that the iron of the electromagnet was extremely soft would make their influence very strongly felt in our results. Then the electromagnet was adjusted to one of the determined azimuths, the torsion angle was read off for the two directions of the field, the current broken, the electromagnet turned to the opposite azimuth, and so on several times. At the end of a series a measurement with one of the first fields was repeated as a control.

Sources of error, difficulties, corrections, and controls.

1. *Inhomogeneity of the magnetic field.* As will be seen from the following discussion this source of error is by far the most important in our case and is indeed the only one that need be taken into account. If we assume that the field near the centre of the pole-gap may be represented by an expression of the form

$$H = H_0 + \frac{r^2}{2}\left(\frac{\partial^2 H}{\partial x^2}\right)_0 (\cos^2\theta - \tfrac{1}{2}\sin^2\theta) \quad . \quad . \quad . \quad . \quad . \quad (3)$$

where H_0 is the field in the centre, and r and θ polar coordinates of a point in the pole-gap with respect to the centre as origin [1]. Let us now replace the ellipsoid by a vertical disc whose diameter is equal to the major axis of the ellipsoid; by taking the expression for the energy of the magnetized disc in the field and differentiating it with respect to the angle between the disc and the lines of force, we obtain for the couple caused by the inhomogeneity of the field:

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[1]) Cf. P. Weiss and H. Kamerlingh Onnes l.c.

$$M' = \frac{3}{16}\, vr^2\, I \left(\frac{\partial^2 H}{\partial x^2}\right)_0 \sin\varphi \cos\varphi . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

($r =$ radius of the disc).

The ratio $\frac{M'}{M}$ of this couple for an angle of 45° to the fundamental couple is

$$\frac{M'}{M} = \frac{3}{16} \cdot \frac{\left(\frac{\partial^2 H}{\partial y^2}\right)_0}{(N_2 - N_1) I} \cdot r^2 .$$

If then we suppose that the relative change of the field in the space occupied by the ellipsoid is of the order of 1 in 1000, the formula given above shews us that although the disturbing couple is a little smaller than the chief couple, the two are *of the same order of magnitude*. Hence we see the great influence that this source of error can have in the investigation of weakly magnetic substances. (With ferro-magnetic bodies it is quite negligible: see the previous paper).

We have accordingly devoted the greatest attention to this source of error. The *conical* pole-pieces were made slightly concave, during which process we every time determined the inhomogeneity of the field by means of a ballistic galvanometer and a small coil that was slightly displaced. We ascertained that the change in the field in a space of about 1 c.c. was certainly less than 1 in 2000. We have not had time to pursue this investigation further, and, besides, we should have to obtain a much more sensitive ballistic galvanometer. But it will be seen that the homogeneity of the field was sufficient for the comparative measurements we proposed to make. We may further remark that all these precautions refer exclusively to the conical pole-pieces; the experiments with the cylindrical pole-pieces were nearly free from these sources of error.

We allow for these disturbing couples in the following way: Assuming that $\left(\frac{\partial^2 H}{\partial x^2}\right)_0 = \lambda H$ the expression for the couple due to inhomogeneity given above becomes ($\varphi = 45°$):

$$\frac{3}{32}\, vr^2\, \lambda\, I H$$

or

$$\frac{3}{32}\, vr^2\, \lambda K H^2,$$

which we shall represent by

$$\beta K H^2 .$$

If α is the angle of torsion of the holder and C the constant of the spring, then

$$C\alpha = \frac{v}{2}(N_1 - N_2)K^2H^2 + \beta KH^2. \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

Thus just as if there were no correction for inhomogeneity the second side of the equation remains always proportional to the square of the field. Even without knowing that correction, if β is itself a constant we should be able to deduce from the observations whether K is a function of the field or not. We see, however, that the constancy of β requires that of λ, i.e. that the field must remain homothetic no matter how great it should be. Now this is not the case as can be seen from the quotients $\frac{2\delta}{H^2}$ in tables V, VII, and VIII. Table V shows first an increase, then the quotient reaches a maximum and diminishes considerably; tables VII and VIII shew a change in exactly the opposite direction; this is just what one would expect if β were variable and K constant, for the tables refer to two practically identical bodies, of which the one is dia- and the other para-magnetic. Now in either case the fundamental couple (uniform field) is in the same direction while the couple due to inhomogeneity changes sign with the susceptibility; should, therefore, the correction in the one case first increase and then decrease, it must in the other case first decrease and then increase. We shall return to this point in § 4.

Since this determination aims only at relative measurements, we have once and for all taken as the value of the susceptibility of oxygen at $-183°$ C. the value that was given by the improved apparatus for measuring the magnetic rise. With the help of this value we have calculated the values of β *for each field* from equation 5): (see tables V and VI). These values fall pretty well on a curve of means. Finally the susceptibility at the lower temperatures is calculated by means of the value of β as a function of the field given by this curve. We shall take the opportunity of the corresponding series of observations to make some remarks upon the influence of the inhomogeneity for each of the three pole-gaps that were used.

2. *The inconstancy of the magnetization as a function of the azimuth.* The general expression for the couple in a uniform field

$$(N_1 - N_2)I^2 v \sin\varphi \cos\varphi$$

only reaches its maximum value just at $\varphi = 45°$, and consequently $\sin\varphi\cos\varphi = {}^1/_2$ since I *remains constant during the torsion.* Here again we see a fundamental difference between the application of this method to the investigation of saturation magnetization and to that of a body of constant susceptibility. It is clear that in the first case the condition $I =$ constant is, as it were, fulfilled by definition.

In our case the deviation from this is by no means *a priori* negligible: the two limiting values of I^2 ($\varphi = 0°$ and $\varphi = 90°$) differ in our case by 0.3 %, and since I^2 always changes between these two limits in the same direction the error caused thereby when $\sin\varphi\cos\varphi = {}^1/_2$ is less than 0.1 %.

In contrast with the two foregoing sources of error, the reaction of the magnetized ellipsoid upon the *distribution of magnetism over the surface of the pole-pieces* can clearly have no effect in the case of a body of small susceptibility while on the other hand, it had to be taken into account in the case of the ferromagnetic bodies. Indeed, with oxygen we have to deal with a magnetization that in the strongest fields of the electromagnet reaches a value of only a few units (in the case of iron it was 1700!).

3. *Influence of the holder.* In this connection we may notice two actions that may go together. In the first place there is the inherent magnetism of the stem, and then there is also an action analogous to that which we wish to measure, for if the stem is not a perfect body of revolution, it is acted upon in the liquid oxygen just as if it were a *supplementary ellipsoid.* We investigated these two sources of error in a blank experiment in liquid oxygen in which the silver ellipsoid was removed, and the surface of the glass was carefully freed from all traces of wax. From this we obtained a maximum of only 1 to 2 ‰ which need not be taken into account

4. *The concentration of the oxygen.* The oxygen in the bath contained a little nitrogen, the concentration of which constantly decreased during the experiment owing to its faster vaporization. So as to be able to allow for this we analysed the gas at the beginning and at the end of each series of observations. The mean concentration was 1.25 % at the beginning and 0.35 % at the end (at the moment that the DEWAR vessel was almost empty). We allowed for this concentration as far as possible; in this respect there remains an uncertainty of about 0.3 %.

5. *Calibration of the suspension springs.* The main torsion spring described above was calibrated outside the apparatus by observing the time of oscillation of a system suspended from it with and without the addition of a known moment of inertia. For the latter we used a bronze ring of rectangular meridian cross section, the diameters and height of which were measured with the cathetometer. Calculation gave the moment of inertia as

582.09 c.g.s.

Care was taken that the spring was subjected to the same tension during the calibration as it experienced while in the apparatus (by attaching suitable weights to it by a torsion-less wire).

For the constant of the spring we found 1184,5 c.g.s.

The platinum-iridium stretching wire gives a torsion couple as well as the spring; the correction for this was determined by the same method as was used in the analogous case by WEISS and KAMERLINGH ONNES (loc. cit.) and it was found to be 0.0152 times the constant of the spring. The difference between the values of the constant at 18° C. and at —190° C. is smaller than the errors of observation. The calculations were therefore carried out with the constant $1184.5\ (1 + 0.0152) = 1202.5$.

6. *Oscillations.* The silver ellipsoid should be protected sufficiently from the influence of oscillations arising from external causes by the occurrence of intensive FOUCAULT currents, but the occurrence of these currents, which were unusually strong gave rise to great difficulties in the observations. In the first place the holder was extremely slow to reach its position of equilibrium. Further, the smallest change in the current flowing through the electromagnet occasioned a sudden kick in the whole moveable apparatus, an immediate result of the oblique position of the ellipsoid with respect to the lines of force. Hence the regulation of the current had to be done with the greatest care. We retained the oil-damper but removed the fixed partitions, for the capillary action of these gave rise to couples that, although small, were still not negligible.

Results of the observations.

TABLE IV*a*.
Cylindrical pole-pieces 21 mm. apart.
$t = -183°.0$ C.

H gauss	double deflection 2δ cm. of the scale	$\frac{2\delta}{H^2} . 10^7$	$K . 10^6$
2250	0.37	0.731	277
4537	1.41	0.685	268.8
6676	3.21	0.7200	275.3
8339	5.10	0.7335	278.1
9387	6.44	0.7307	277.5
10120	7.45	0.7274	277.0
10685	8.26	0.7234	276.1
11130	8.99	0.7258	271.6
11440	9.38	0 7167	274.8
11765	9.90	0.7155	274.6

Mean $K_{90°.1K.} = 275.6$

TABLE IV*b*.
Cylindrical pole-pieces 21 mm. apart.
$t = -201°.1$ C.

H gauss	double deflection 2δ cm. of the scale	$\frac{2\delta}{H^2} . 10^7$	$K . 10^6$
2250	0.50	0.988	324
4537	2.10	1.020	328.0
6676	4.66	1.041	331.1
8339	7.36	1.058	334.0
9387	9.17	1.042	331.4
10120	10.63	1.038	330.8
10685	11.81	1 034	330.2
11130	12.70	1.025	328.6
11440	13.41	1.024	328.4
11765	14.14	1.022	328.0

Mean $K_{72°.0\text{K.}} = 330.0.$

Without being corrected for lack of uniformity in the field the means give the following values:

$$\chi_{90°.1\text{ K.}} = \frac{275.6}{1.143} . 10^{-6} = 241.1 . 10^{-6}$$

$$\chi_{72°.0\text{ K.}} = \frac{330.0}{1.230} . 10^{-6} = 268.3 . 10^{-6}.$$

The corresponding results obtained by the method of the magnetic rise were

$$240.6 . 10^{-6} \text{ and } 269.3 . 10^{-6}.$$

The differences between the results as obtained by the two methods are scarcely 0.4 %. This gives us great confidence in the ellipsoid method even for this particularly difficult determination, and it shews that the method is also suitable for absolute measurements if only the necessary care is taken to ensure the uniformity of the field and the correctness of the shape of the ellipsoid.

We must remember that there was a great number of absolute

measurements whose results had to be used (axes and volume of the ellipsoid, constants of the springs, magnetic field, density of the liquid oxygen) and also that the shape of the ellipsoid was not perfect. On the other hand we must remark that the application of the correction for the non-uniformity of the field might conceivably have diminished the correspondence between the results obtained by the two methods. We have, however, both theoretical and experimental grounds for the assumption that this correction remains within the limits of accuracy of not more than 0.5% in the case of cylindrical pole-pieces with flat end surfaces 90 mm. in diameter and at a distance of 21 mm. apart.

TABLE V*a*.
Conical pole-pieces 20 mm. apart.
$t = -183°.0$ C.
(To determine β we assumed $\chi_{90°.1} = 240.6 . 10^{-6}$).

H gauss.	Double deflection 2δ cm. of the scale.	$\frac{2\delta}{H^2} . 10^7$	$10^6 \beta$
3685	1.27	0.935	58.5
4615	1.96	0.920	54.7
6944	4.55	0.9437	60.8
9205	7.96	0.9400	59.7
11280	11.90	0.9348	58.5
12835	15.44	0.9374	59.1
14015	18.26	0.9295	57.0
14900	20.19	0.9098	51.9
15585	21.73	0.8945	48.1
16120	22.87	0.8802	44.5

A graph of β as function of H was made, which was used for the following table.

TABLE V*b*.
Conical pole-pieces 20 mm. apart.
$t = -208°.2$ C.

H gauss.	Double deflection 2δ cm. of the scale.	$\frac{2\delta}{H^2} . 10^7$	$\beta . 10^6$	$K . 10^6$
2296	0.79	1.498	56.0	[357]
4615	2.75	1.291	57.8	[328]
6944	6.82	1.414	58.5	344.2
9205	12.15	1.435	59.8	346.3
11280	18.25	1.434	59.7	346.2
12835	23.67	1.437	59.2	346.9
14015	27.89	1.420	56.5	346.1
14900	30.84	1.389	52.4	345.1
15585	33.24	1.368	48.0	345.0
16120	35.44	1.363	43.8	347.3

The mean with the exception of the two values placed between brackets is 345.9 and it gives

$$\chi_{64°.9\,\mathrm{K.}} = 275{,}0 \,.\, 10^{-6}$$

while the method of the magnetic rise gave

$$\chi_{64°.9\,\mathrm{K.}} = 283{,}5 \,.\, 10^{-6}$$

The difference is 3°/₀; but in this connection we must remember that the correction for non-uniformity is about 16°/₀, and that the temperature of the liquid becomes very uncertain at the pressure of 11 mm. under which the liquid boils at this temperature.

Finally, we now give two series of measurements which were made with other pole-gaps so as to obtain other deviations in the uniformity of the field. They were hastily made and under unfavourable circumstances, since oscillations and disturbances caused by the running of machines in the neighbourhood interfered with the observations. We give them more as examples of how the method of calculation followed still leads to good results even when the couples due to non-uniformity of the field are extremely large (28°/₀ of the chief couple).

TABLE VI*a*.
Conical pole-pieces 18.2 mm. apart.
$t = -183°.0$ C.
(To determine β we assumed $\chi_{90°.1} = 240.6 . 10^{-6}$).

H gauss	double deflection 2δ cm. of the scale	$\frac{2\delta}{H^2} . 10^7$	$\beta . 10^6$
5013	3.34	1.328	159.2
7547	7.31	1.283	147.4
9993	12.64	1.246	137.9
12165	18.33	1.238	136.2
13760	22.39	1.183	121.7
14900	26.26	1.182	121.5
15750	28.83	1.162	116.6
17005	35.58	1.230	133.9

β was again graphed as a function of H, which led to the correction for K in the following table.

TABLE VI*b*.
Conical pole-pieces 18.2 mm. apart
$t = -208°.2$ C.

H gauss	double deflection 2δ cm. of the scale	$\frac{2\delta}{H^2} . 10^7$	$\beta . 10^6$	$K . 10^6$
5013	46.8	1.861	152	341
7547	10.71	1.880	146.5	348
9993	18.88	1.862	137.5	351
12165	27.92	1.885	130.7	357
13760	35.35	1.868	125.5	359
14900	41.36	1.861	123.0	360
15750	45.67	1.840	122.5	357
17005	51.81	1.791	127.5	347

The mean 353 gives $\chi = \frac{K}{1.255} = 279.5.10^{-6}$ a value that is not much smaller than $283.5.10^{-6}$, which was obtained by the method of the magnetic rise.

Solid oxygen.

§ 4. *Ellipsoid of solid oxygen.* In this case observations had to be made directly upon an ellipsoid of oxygen. The oxygen therefore had to be frozen in a mould of approximately the same form and dimensions as the solid silver ellipsoid described above. This new condition necessitated the following experimental arrangement.

The cover and the DEWAR tube are the same as for liquid oxygen, with the exception of the cap D. The adjusting tube is also the same, but it is so arranged that it can be moved as a whole up or down, while the whole apparatus remains closed and in its place. With this end in view it is attached to the tube m, which moves through the stuffing-box D''_2; this corresponds to D_2 of the liquid oxygen apparatus, but in this case the wide glass tube C_1 is lengthened by a rigid brass tube M that serves to give sufficient play to the vertical movement of the whole adjusting tube. The former stem k had to be lengthened by the same amount (L''_1 L''_2), and is contained in the tube m.

The holder is also a glass tube b''; it is not however closed, but at b''_3 it changes into a very much narrower tube (0.5 mm.) that ends at b''_1 in a glass ellipsoid a''. To this ellipsoid there is fused a solid stem b''_5 that connects it with the stretching wire. The oxygen gets to the ellipsoid through the holding tube which it enters at b''_6. A rubber tube n ($d = 3$ mm.) admits the gas from outside; it is attached to the inlet tube n_4 that passes through the cover and is soldered to it. With this arrangement it is easy to cause the oxygen to solidify inside the ellipsoid. When the apparatus is ready for use the adjusting tube is pulled upwards by the cap Δ till the glass ellipsoid reaches the unsilvered part of the vacuum glass. The vacuum glass is then filled with liquid hydrogen. While the ellipsoid is still connected with a reservoir of oxygen, the adjusting tube with the ellipsoid is slowly pushed downwards until it does not quite touch the liquid hydrogen but is in its vapour. The oxygen is then seen to condense slowly, and, if the operation is carefully performed, the whole ellipsoid and supply tube are seen to fill with liquid oxygen. The tube being lowered still further, vapour is reached that is sufficiently cold to cause the oxygen to solidify. On account of the large

contraction of the oxygen on solidification it is seldom that one does not see some empty space in the ellipsoid; the operation must then be repeated several times, since the oxygen that is still liquid at this temperature has a pretty great viscosity and flows with difficulty from the tube; we shall return to this point later. When the ellipsoid is completely filled with solid oxygen the adjusting tube may be lowered right down. A mark is made beforehand, so that the ellipsoid may be accurately adjusted to the centre of the gap when the silvered tube is again in its place.

Errors, corrections, auxiliary measurements.

1. *Couples due to inhomogeneity.* As will presently appear, we made measurements not only in liquid hydrogen (solid oxygen), but also keeping everything else the same, at two temperatures in a bath of liquid oxygen (i.e with the same ellipsoid of liquid oxygen). Since the susceptibility of the liquid oxygen was known, we had therefore two measurements of the couples due to inhomogeneity as a function of the field; they are given in Table VII. As a result of the somewhat smaller dimensions of the ellipsoid, these corrections are comparatively much less important.

2. *Purity of the oxygen.* The oxygen was freed from nitrogen by vaporizing a large quantity of impure liquid oxygen under reduced pressure.

3. *Density of the solid oxygen.* We have already mentioned the difficulty of completely filling the ellipsoid with solid oxygen. On account of the opaqueness of the oxygen that has already solidified one cannot with certainty assert that this condition has been fulfilled [1]).

Since the specific susceptibility is determined from a known volume this error would have immediate effect upon the result. We tried to eliminate this error as well as possible by determining the density with the same ellipsoid by filling it with solid oxygen under the same circumstances as those obtaining in the experiments and then measuring the quantity of gas formed from it on vaporization. We may assume that the small cavities that may form are pretty much the same in the various cases. Indeed, from two similar measurements the density measured in this way was found to vary by only about $1^0/_0$. By taking as the mean density that determined by these experiments, the eventual presence of cavities is allowed for. In this way we obtained

$$\varrho = 1.41.$$

The *absolute* values of the couples due to inhomogeneity of the

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1) When there is an empty space af a few mm^3, however, it can be seen quite well.

field are not modified by a cavity formed in the vertical axis, as was usually the case, for it is clearly those portions towards the surface of the ellipsoid that are the chief contributors to them. On the other hand, they might obtain a greater *relative* influence, but as the observations shew, the sum of the corrections arising from this cause is so small that they may be regarded as independent of the susceptibility within the limits of accuracy of the experiments. In that case this difficulty completely disappears.

4. *Dimensions of the ellipsoid.* The internal volume was obtained by filling the ellipsoid with mercury and weighing it. It was 0.1812 c.c. The change of volume under atmospheric pressure was found to be of no account by pumping the space above the mercury free from air and observing the position of the mercury in the capillary.

The external axes were measured directly. Then the thickness of the glass at ten different points was determined by focussing a microscope on the image of the outer surface formed on the mercury with which the ellipsoid was filled. It changed but slightly from place to place. The mean was taken and twice that value was subtracted from the external measurements. The results were:

1.044 cm.

and

0.335 cm.

Calculating the volume from these figures we get 0.1925 c.c. which is about 6 % greater than the true volume as directly determined. This is accounted for by the special shape of the meridian section which curves somewhat too strongly at the outer ends. For calculating the coefficients of demagnetization we took a mean ellipsoid with the same major axis and the minor axis small enough to give the real volume[1]). The data for the calculation were therefore:

1.044 cm.

and

0.3173 cm.

5. *Opposing couple.* The suspension spring and the stretching wire were the same as were used for the liquid oxygen. We must, however, allow for the rubber supply tube for the oxygen. This (which was chosen as thin as possible) modified both the zero and the constants of the total opposing couple, as soon as the pressure

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[1]) It is clearly not quite right to do this; there are, however, experimental data to support this method of correcting: V. QUITTNER (Diss. Zürich 1908, also Arch. sc. phys. et nat Genève, Sept.—Nov. 1908) found that this method of treatment was sufficiently accurate even for discs, bodies that deviate far more from an ellipsoid than those we used.

difference between the inside and the outside of the tube appreciably altered (on account of the change in shape of the tube). In all our experiments, therefore, we took care that there was a constant pressure difference of 70 mm. between the pressure inside the cover and that inside the holder (the latter was the smaller of the two). We got a very sensitive indication of the constancy of this difference not only from the manometers but also from the zero position of the holder. Experiments carried out outside the apparatus shewed that the constant of the total couple changed about 10°/₀ between the complete flattening of the rubber tube by the atmospheric pressure and equality between the pressures on both sides. This corresponds to a deflection on the reading scale of *more than a metre*. If we assume rough proportionality we find that a displacement of 1 cm. would indicate a change in the opposing couple of only 0.1 °/₀. The zero was kept constant to a few millimetres.

The calibration was made under circumstances exactly the same as in the experiments (pressure difference, etc.).

The total constant with the addition of that of the stretching wire was

$$1503 + 18 = 1521 \text{ cgs.}$$

Results.

TABLE VII.

Calculation of the corrections for non-uniformity from observations made in a bath of liquid nitrogen.
Conical pole-pieces 20 mm. apart.

H gauss	$t = -195°.6$ C.			$t = -210°.0$ C.			$\beta.10^4$ (mean). Double weight given $-195°.6$
	2δ cm.	$\frac{2\delta}{H^2} 10^7$	$\beta.10^4$	2δ cm.	$\frac{2\delta}{H^2} 10^7$	$\beta.10^4$	
4615	1.18	0.554	—0.137	1.59	0.746	—0.199	—0.158
6944	2.69	5577	127	3.53	0.7322	232	162
9205	4.73	5580	126	6.21	7330	231	160
11280	7.08	5560	132	9.23	7251	250	171
12835	9.17	5564	131	12 10	7341	228	163
14015	11.14	5670	100	14.49	7378	219	140
14600	12.90	5812	060	16.94	7613	162	094
15585	14.29	5884	039	—	—	—	—
16120	15.67	6031	003	20 21	7781	121	040

It can be seen that the values obtained for β are not the same at the two temperatures. Meanwhile it has to be applied here only as a correction for the susceptibility of solid oxygen which at the most is 3%. A difference of temperature of $1°$ C. in the bath under reduced pressure gives more than half the difference between the two values, whence we have given the determination under reduced pressure only half the weight accorded the measurement at ordinary pressure.

The uncertainty of the mean has less than 1% influence upon the value of the susceptibility of solid oxygen. The curve for β as well as its sign correspond with what were found for the silver ellipsoid.

TABLE VIII.
Susceptibility of solid oxygen.
$t = -252°.8$
(bath of liquid hydrogen boiling under atmospheric pressure).

H gauss	2δ cm. of the scale	$\frac{2\delta}{H^2} . 10^7$	$K . 10^6$ uncorrected	$K . 10^6$ corrected according to tab. VII.
2296	0.89	1.69	519	533
4615	3.57	1.676	518.3	532.9
6944	7.92	1.642	512.3	527.3
9205	14.07	1.660	515.0	530 1
11280	21.14	1.661	515.2	530.1
12835	27.92	1.684	518.7	532.8
14015	32.96	1.678	517.6	529.0
14900	37.38	1.683	518.4	527.5
15585	40.77	1.678	517.6	523.7
16120	44.05	1.696	520.0	523.6

Mean 529.0

whence it follows that $\chi_{20°.3\,\mathrm{K.}} = \frac{529.0}{1.41} 10^{-6} = 375.2 . 10^{-6}$.

TABLE IX.
Susceptibility of solid oxygen.
$t = -258{,}9°$
(bath of liquid hydrogen under 70 mm. vapour pressure).

H gauss	2δ cm. of the scale	$\frac{2\delta}{H^2} \cdot 10^7$	$K.10^6$ uncorrected	$K.10^6$ corrected
2296	1.19	2.257	600.5	614.5
4615	4.80	2.253	600.1	614.6
6944	10.86	2.252	600.0	615.0
9205	19.24	2.270	602.2	617.1
11280	28.13	2.210	594.3	609.2
12835	37.09	2.250	599.6	613.7
14015	45.05	2.293	604.7	616.7
14900	51.24	2.306	606.8	615.9
15585	55.73	2.293	605.3	611.1
16120	60.20	2.317	608.5	612.1

From the mean 614.0 follows $\chi_{sol.\,14°.2K.} = \frac{614.0}{1.41} = 435.6 \,.\, 10^{-6}$. The products into $\sqrt{T}$ are

$$-252.°8 \qquad 375.2 \,.\, 10^{-6} \sqrt{20.3} = 1690 \,.\, 10^{-6}$$
$$-258.°9 \qquad 435.6 \,.\, 10^{-6} \sqrt{14.2} = 1641 \,.\, 10^{-6}.$$

Hence we can represent the two observations pretty well by

$$\chi_{sol.} = \frac{1690}{\sqrt{T}} \,.\, 10^{-6},$$

which is adjusted to the measurement at the higher temperature.

The deviation from this ratio for the lower temperature, however, is somewhat greater than the errors of observation.

§ 5. *Summary and conclusion.* As regards the dependence of specific susceptibility upon temperature our most reliable determination gives

$$\chi_{liq.\,90°.1\,K.} = 240.6 \,.\, 10^{-6}.$$

CURIE found $\chi = \frac{33700}{T} \,.\, 10^{-6}$ between 20° C. and 450° C. whence it

would follow that for $T = 90°.1$ K. $\chi = 374 . 10^{-6}$ a number that differs essentially from ours [1]).

There is therefore no possibility of extrapolating CURIE's law to the liquid phase of oxygen. This was also the conclusion reached by FLEMING and DEWAR in their first treatment of the question, but after more careful experiments they rejected their former result [2]).

The results obtained from the two magnetic rise apparatus at lower temperatures can, within the limits of experimental error, be expressed by a very simple law: *the specific susceptibility is inversely proportional to the square root of the absolute temperature.* From the observations obtained with the more reliable apparatus we deduce the formula

$$\chi_{liq.T} = \frac{2284}{\sqrt{T}} . 10^{-6}$$

which holds to within 5%. None of the results obtained by the method of the maximum couple are in conflict with those deduced from the formula.

The results with solid oxygen approximately follow the relation

$$\chi_{sol:1} = \frac{1690}{\sqrt{T}} . 10^{-6}.$$

At the lowest temperatures there is a small deviation indicating a smaller increase at lower temperatures; it is, however, so small that we may still accept the formula given as approximately correct for the solid state of aggregation below the melting point of oxygen and down to 14°.2 K.

Further experiments at more numerous temperatures must show exactly how far these deductions hold for the liquid and solid states. They shew (see fig. 5) that there is a jump in the value of χ at the melting point, since

$$\chi_{liq.T_m} = 1{,}3\ \chi_{sol.T_m}$$

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[1]) R. HENNIG's (1893) result should give

$$\chi = \frac{27600}{T} 10^{-6} \text{ and } \chi_{90°.1\text{ K.}} = 307 . 10^{-6}.$$

[2]) FLEMING and DEWAR's results: 1st paper (1896) $\chi_{90°.1K.} = 200 . 10^{-6}$; 2nd paper (1898) $28i . 10^{-6}$, mean $243.5 . 10^{-6}$ pretty much the same as our result. According to the mean of the result of FARADAY and BECQUEREL the specific susceptibility for oxygen at 0° is $91 . 10^{-6}$; this gives by extrapolation from CURIE's law $\chi_{90°.1K.} = 299 . 10^{-6}$. The English savants used this number in their second research for the comparison of the susceptibility of liquid oxygen with that of the gas.

We hope to answer the question if this jump really exists by special experiments arranged for the purpose; we may, in the meantime, consider that it does probably exist. What CURIE found in the transformation of γ iron to δ iron is analogous to the sudden change which we here assume to exist while the form of the law remains unaltered, and which can occur at the melting point or at a point of transformation to an allotropic modification. WEISS [1]) has shown that this can be accounted for on the assumption that at this particular point di-atomic iron changes into tri-atomic.

On the other hand we consider it probable that the law according to which the specific susceptibility increases with the temperature, viz: inverse proportionality to the square root of the absolute temperature at lower temperatures, gradually transforms into that of inverse proportionality (CURIE's law) at higher temperatures, and that each of these laws, therefore, may be but approximative to the same function over different ranges of values of the independent variable T.

The supposition that the change of specific susceptibility with density is of no importance lies at the bottom of the assumption of the gradual transformation of CURIE's law into that of $T^{-\frac{1}{2}}$. If, on the other hand, we assume that this change is of importance, that e. g. when the internal pressure is considerable the molecules under its influence undergo not only a compression but also a lessening of their magnetic moments, then a region of great molecular compressibility in which the specific susceptibility should change both with the temperature and with the density should exist between the gaseous phase in which the specific susceptibility would be pretty well independent of the pressure, and the liquid phase at lower temperatures, in which the molecules would not be appreciably affected by an additional external pressure on account of their already great internal pressure, and in which, therefore, the specific susceptibility would also be pretty well independent of the pressure. As regards the difference between the magnetic moment of the elementary magnets in the condition of saturated liquid and vapour and that at normal or smaller density at the same temperatures, it is to be expected according to that representation, that this difference will change with temperature in consequence of the change of density with temperature.

The assumption can also be made that complex molecules are formed in the liquid state, and that these diminish the intensity of the elementary magnets; in that case changes in susceptibility of

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1) P. WEISS, loc. cit.

mixtures of liquid oxygen with non-magnetic gases should obey the thermodynamic laws that govern the number of such complexes. But all this must be established by further experiments which we hope to complete; in the meantime the most probable assumption is the old one that the specific susceptibility is independent of the pressure.

As regards the question as to whether the specific susceptibility at lower temperatures still follows the law of inverse proportionality to the root of the absolute temperature, if the ferro-magnetism with a very low-lying CURIE point according to WEISS's theory of corresponding magnetic states does not exist, then the change to a still slower increase with decreasing temperature and the approximation to a limiting value is, perhaps, more probable.

The law of $T^{-\frac{1}{2}}$ at once gives rise to the question if instead of the LANGEVIN elementary magnets whose intensity is independent of the temperature, we should assume that their intensity varies directly as $\sqrt{T}$; that is, that we should assume the existence of elementary currents or electrons moving in their paths with speeds proportional to (and, therefore, determined by) the speeds of molecular heat motions. In other words, while LANGEVIN's theory already supposes that the planes in which the electrons move follow the motions of the molecules, but that the areas described in those planes are still independent of heat motion, we should now assume that the electrons undergo the influence of heat motion at their motion *in* their paths, and, if the radius of their path has also become invariable, revolve while remaining in the same position with respect to the atom; they would be electrons that are frozen fast to the atom, an assumption that has already been made to explain other phenomena.

This addition to LANGEVIN's theory, however, does not lead to a specific susceptibility proportional to $T^{-\frac{1}{2}}$ as one at first sight would be inclined to think, but to a constant specific susceptibility.

To substantiate that addition it will probably be necessary to proceed to still lower temperatures than those of our experiments. It seems at present that it is not impossible that then the law χ proportional to $T^{-\frac{1}{2}}$ changes to $\chi = const.$: our observations on solid oxygen seem to indicate a change in this direction. The assumption to which this is equivalent: viz, that the magnetic motions of the electrons cease at the absolute zero, and to which our experiments seem to lead, is much more satisfactory than that the magnetic motions of the electrons still persevere even at the absolute zero.

The second question to which we devoted attention — the dependence of susceptibility upon field strength requires no detailed treat-

Fig 1.

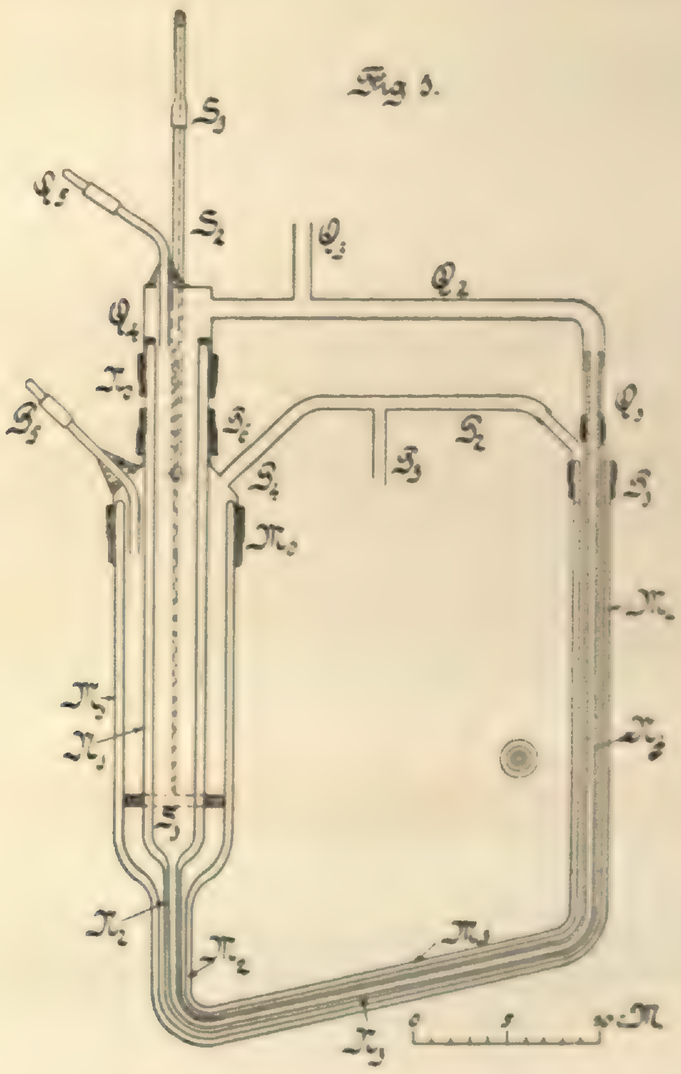
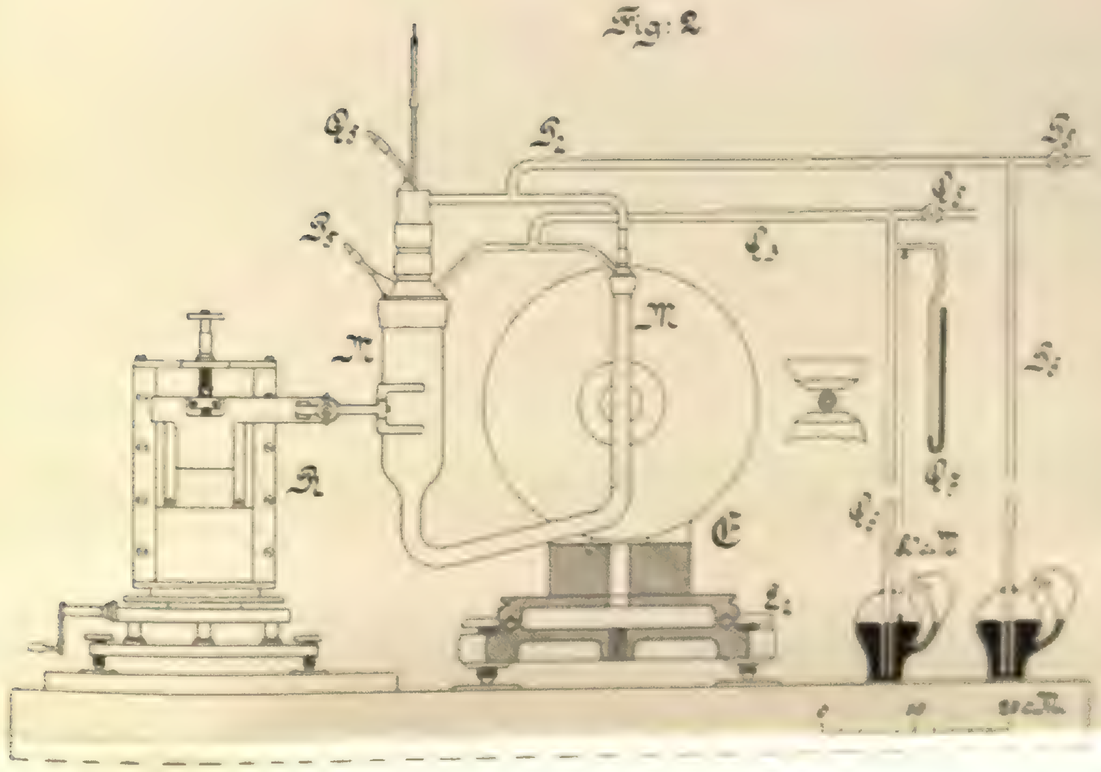
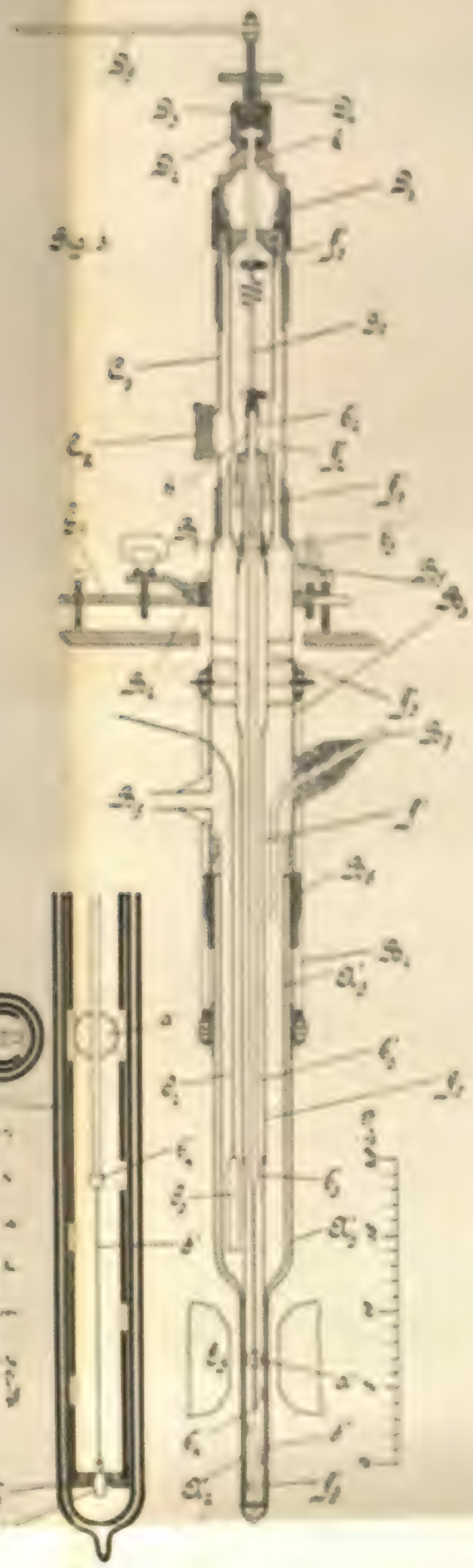


Fig: 2





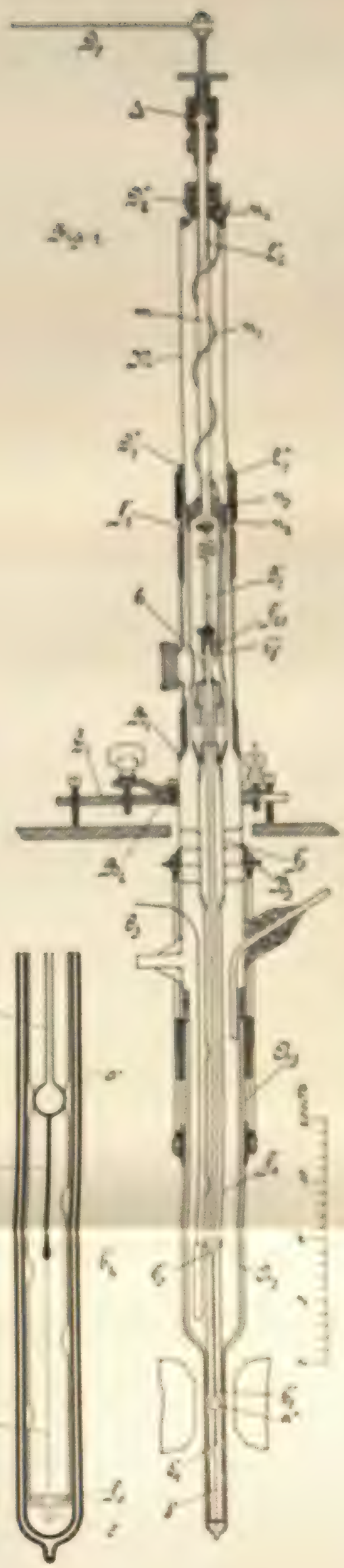
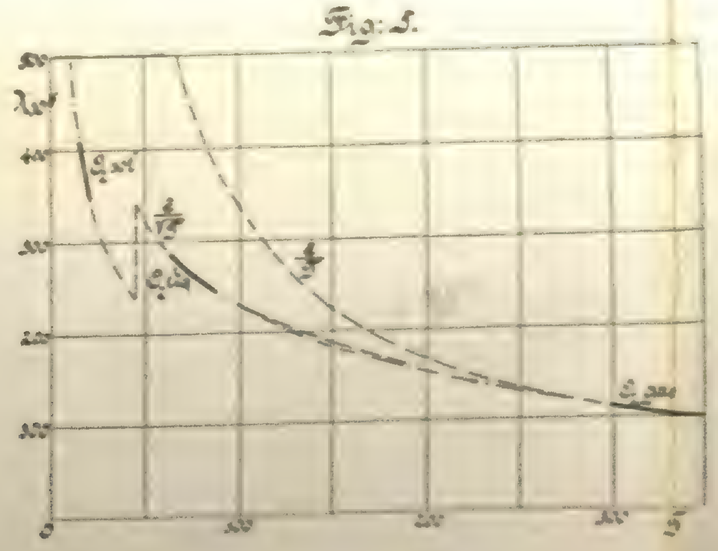


Fig: 3.



ment. The method of the magnetic rise seemed in some instances to give a decrease of the order of 1% in a field of 8000 gauss, while the method of the maximum couple gave with the cylindrical pole-pieces up to 12000 gauss only a very small systematic deviation and with the conical pole-pieces (16000 gauss) the deviation was scarcely appreciable.

The solid oxygen ellipsoid with which a much lower temperature was reached seemed to give a small decrease at 16000 gauss; it is possible, however, that a greater deviation is obscured by the correction for the non-uniformity of the field. We consider, however, that, assuming that the experiments were accurate to within 1% the change of the susceptibility with the field up to 16000 gauss remains within the limits of experimental error. This is in agreement with the theory of LANGEVIN, if this, notwithstanding the deviation from CURIE's law, is still applied.

Physics. — "*The magneto-optic* KERR-*Effect in ferromagnetic compounds and alloys*". By STANISLAW LORIA. (Communication from the Bosscha-Laboratory).

It has been shewn by KAZ [1]), RIGHI [2]), KUNDT [3]), SISSINGH [4]), ZEEMAN [5]) and also by KERR [6]) himself that the phenomenon discovered by the last named in 1876 depends not only on the orientation of the reflecting surface with respect to the magnetic vectors, but also (in a somewhat complicated manner) on the angle of incidence and the position of the plane of polarization of the incident beam. In the simplest and by far the most important case of almost normal incidence of light polarized perpendicularly or parallel to the plane of incidence, the reflected light in general is elliptically polarized according to RIGHI [7]); the rotation of the major axis of the ellipse depends on the magnetisation and the wave-length.

According to the measurements made by DU BOIS [8]) it is in every case proportional to the former; as regards the variation with the

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1) P. C. KAZ, Diss., Amsterdam 1884.
2) A. RIGHI, Ann. de Chim. et Phys. (6) **4** p. 433, 1885.
3) A. KUNDT, Wied. Ann. **23** p. 228, 1884; **27** p. 199, 1886.
4) R. SISSINGH, Arch. Néerl. (1) **27** p. 173, 1894.
5) P. ZEEMAN, Leiden Comm. no. 15, 1895; no. 29, 1896. Arch. Néerl. **27** p. 252 1894.
6) J. KERR, Phil. Mag. (5) **3** p. 339, 1877. Phil. Mag. (5) **5** p. 161, 1878.
7) A. RIGHI, Ann. de Chim. et Phys. (1) **9** p. 120, 1886.
8) H. DU BOIS, Wied. Ann. **39** p. 25, 1890.

latter, the rotatory dispersion, according to the same author, shews certain regularities. For iron, cobalt, and nickel the rotations visually observed were always *negative*; for iron the dispersion-curve seems to indicate a numerical minimum in the ultraviolet and thence ascends from violet towards red; in the case of cobalt the minimum occurs between blue and green, and for nickel in the yellow. These numerical minima of negative rotation may be considered *algebraic* maxima, their wave-length increasing as the metal's position in the periodic system advances. For magnetite the observed rotations were in every case *positive*, though the curve appeared directed towards negative values beyond the blue; a distinct maximum occurred in the yellow, corresponding to the above algebraic maxima.

More recently INGERSOLL [1]) has contributed important papers relative to this subject; he was able to supplement DU BOIS' curves in the infra-red up to about $3\,\mu$. According to this author the complete rotatory dispersion-curves thus obtained shew a marked resemblance to a typical dispersion-curve in the region of an exceedingly broad band of resonance-absorption. The particular cases of nickel and magnetite are notable, for the rotation appears to vanish between 1 and $1{,}5\,\mu$ and then to change in sign.

Further progress in this subject was difficult in view of the fact, that as yet the only ferromagnetic substances suitable for a study of the KERR-Effect were the four above-mentioned bodies. Several attempts to study with reflected and transmitted light the magneto-optic phenomena connected with the KERR-Effect were made with partially transparent films of metals prepared electrolytically, after the manner of KUNDT, or by cathodic discharge. Although the latest investigations [2]) on the optical properties of these films of magnetic metals brought to light further interesting but confusing results, yet the conditions in the films can obviously depend on their structure and on their mode of preparation in a very complex way. As a reflecting surface such a film is certainly inferior from a physical point of view to a mirror polished on a compact and massive block of metal.

An attempt to add to the number of substances which exhibit the KERR-Effect was thus of some interest. I entertained some hopes in this respect, since several chemists of late have synthetically prepared new ferromagnetic substances. In the first place, a number of combinations of different oxides with iron oxide, so called metaferrites,

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[1]) L. R. INGERSOLL, Phil. Mag. (6) **11** p. 41, 1906 & **18** p. 74, 1909.

[2]) C. A. SKINNER & A. Q. TOOL, Phil. Mag. (6) **16** p. 833, 1908. H. BEHRENS Inaug. Diss. Münster i. W. 1908. L. R. INGERSOLL, loc. cit.

prepared by HILPERT[1]), presented an interesting field of research. In all these cases, the chemical structure resembles that of ferroferrite (ferroso-ferric oxide), in that the iron sesquioxide plays the acidic part, thus imparting ferromagnetic properties to the compound. Of this class of substances however, only cupriferrite and calciumferrite could be obtained in a state suitable for my experiments. Secondly, certain alloys of more or less ferromagnetic metals, and in particular those of nickel-iron[2]), together with the well-known ternary HEUSLER alloy, and WEDEKIND's[3]) binary manganese-antimony alloy present considerable interest. So far as I am aware, the magneto-optic properties of these alloys have been only partially investigated, the only account of similar experiments, which I have come across, being INGERSOLL's communication previously referred to and a Russian paper by TOKMATSCHEW[4]), who described experiments with HEUSLER's alloy.

I have studied the magneto-optic properties of the above mentioned bodies and also those of the well-known magnetic chromic oxide. Below an account of the preliminary results of my research is given.

Experimental Arrangement. Solar rays were exclusively used; they passed through a direct-vision monochromatic illuminator[5]), with divergence 1:4, thus furnishing light of great intensity. The rays passed (Fig. 1) through a lens (L), a total reflecting prism (P), a LIPPICH's arrangement of two halfshade Nicols (N_1, N_2) and falling nearly normal on a mirror between the two poles of an electromagnet, were reflected, finally passing through an analyser (N_3) and a telescope of fourfold magnifying power. The dimensions of the lenses, of the diaphragms, of the width and angle of the conical bores in the cores and poles were all calculated beforehand, particular care being taken to maintain maximum brightness, a uniform field of view, and also the avoidance of all unnecessary reflections[6]).

The observations were carried out with nearly normal incidence. RIGHI[7]) found, that up to an angle of incidence of 15° there was

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1) S. HILPERT. Ber. deutsch. Chem. Ges. **42** p. 2248, 1909. Verh. deutsch. Phys. Ges. **11** p. 293, 1909.

2) CH. ED. GUILLAUME, Les aciers au nickel, Paris 1898.

3) E. WEDEKIND, Ztschr. f. phys. Chem. **66** p. 614, 1909. K. HONDA, Ann. d. Phys. **32**, 1910.

4) S. TOKMATSCHEW, Journ. d. russ. phys.-chem. Ges., **42** (phys. T.) p. 15, 1910.

5) H. DU BOIS, Verh. d. D. Phys. Ges. **11** p. 708, 1909.

6) A description of the analyser and polariser mentioned is given by H. DU BOIS, Wied. Ann. **46**, p. 545, 1892.

7) A. RIGHI, Ann. de chim. et de phys. (1) **9** pp. 120, 132, 1886.

scarcely any variation of the effect. However, in my experiments the angle between the incident and reflected beams was only 2° or 3°. The incident light was polarized horizontally in the plane of incidence. From RIGHI's observations it is known that even a normal incident beam of linearly polarized light when reflected from a magnetized mirror becomes elliptically polarized, the ellipticity however being only slight; ZEEMAN [1]) later measured this ellipticity in the case of iron and cobalt. Up to the present the evaluation of the ellipticity in my experiments has not been attempted; I considered that the slight reflecting power of some of my mirrors would not warrant such an attempt, and in addition it must be borne in mind, that the rotations themselves are small. Moreover the ellipticity, if any, must be nearly inappreciable, for by employing the best of my mirrors and by carefully avoiding diffused light, I have never been confronted with any difficulties, while the extinction of light in each half of the field of view was satisfactory. Even when the rotations are very small it is possible by means of the half-shade arrangement to observe and to measure them with sufficient accuracy. Hence it was thought unnecessary to use the method of multiple reflections, thereby avoiding new complications and further sources of error. The azimuth of the analyser was determined by means of a vertical scale seen through a combination of mirrors.

For the production of the magnetic field a small DU BOIS semicircular electromagnet of resistance 9 Ω was employed. To avoid the danger of sparking with reversal of current about 60 Ω were shunted across its terminals. The field was determined by means of a standardised thin glass-plate silvered at the back, which could be placed immediately in front of the mirror. The light ($\lambda = 589\,\mu\mu$), being reflected by the mirror as described above, suffers a double magnetic rotation in the glass. The ensuing very slight double rotation of the light in its passage to and fro through the magnetized air could be computed from the data of SIERTSEMA [2]), but proved quite negligible. Indeed, by using a silver-mirror, it was found that the rotation lies within the limits of experimental error. All the measurements were made with "polar" magnetization and at ordinary temperature.

Test-Specimens. The following substances were experimented upon: *Cupriferrite* ($CuO . Fe_2O_3$), *Calciumferrite* ($CaO . Fe_2O_3$), *Magnetite* (Ferroferrite) ($FeO . Fe_2O_3$), *Ilmenite* ($Ti_2O_3 . Fe_2O_3$), ferromagnetic *chromic oxide* (Cr_5O_9), "*Invar*" (36 Ni, 64 Fe), the HEUSLER

[1]) P. ZEEMAN, Leiden-Comm. No 15, 1895.

[2]) L. H. SIERTSEMA, Versl. Kon. Akad. Wet. Amsterdam 7 p. 289, 1899.

alloy (26 Mn, 13 Al, 61 Cu). The first two were kindly prepared by Dr. HILPERT in the metallurgical Laboratory of the "Technische Hochschule" in Charlottenburg; the natural magnetite is from the collection of the Bosscha-Laboratory, and is the same specimen, possessing a polished octahedral surface, which was formerly examined by DU BOIS [1]). A very fine-formed crystal of ilmenite was kindly lent by Prof. LIEBISCH. The HEUSLER alloy was supplied by the DE HAËN chemical factory in Seelze; its interior was full of bubbles, but its surface was capable of polish and supplied a very good mirror. The "invar" contained about 36 % Nickel and came from France (Société de Commentry-Fourchambault). For the chromic oxide I am indebted to Dr. KOPPEL. I desire to express my obligations to all the above mentioned gentlemen.

Throughout this paper I shall denote as usual by: $\mathfrak{H}$, the field intensity in kilogausses, $\mathfrak{J}$ the magnetization, $\mathfrak{J}_m$ its saturation value, ε single rotation of the plane of polarization in minutes, K, KERR's constant. In the tables, the column under N shows the number of readings in each series of measurements, which depended upon the polish of the mirror and the variable brightness, λ denotes the wave-length in $\mu\mu$, Δ the direct scale-reading in mm. of the double rotation produced by reversal of the current. The average values of the single rotations are given in the fourth column and in the fifth and sixth the average errors in minutes and in percentages.

The sense of the rotation is referred as usual to that of the magnetizing current; e.g. in the case of iron the "polar" KERR rotation is negative.

Results. The results obtained with the various substances were as follows:

1. *Cupriferrite.* Measurements were made on two mirrors of this material with similar results. The relation between the rotation and the wave-length in a field of 10,2 kgs. is shown numerically in Table 1 and graphically in Fig. 2. The dispersion-curve exhibits a type which has not been observed hitherto in the visible spectrum.

In the violet the rotation is *positive*, a maximum occurring in the blue; with increasing wave-length the rotation gradually decreases and in the neighbourhood of 587 $\mu\mu$ goes through zero, becoming *negative* for longer wave-lengths. Between 640 and 670 $\mu\mu$ a rather flat minimum is exhibited, the curve then gradually proceeding upwards. The rotations are small throughout, the maximum value not being

[1]) H. DU BOIS, Wied. Ann. **39**, p. 25, 1890.

TABLE 1.

ε = funct (λ) Cupriferrite $\mathfrak{H}$ = 10.25 Kgs.

N	$\lambda(\mu\mu)$	Δ (mm)	ε (Minutes)	$\delta\varepsilon$
19	436	+ 8.3	+ 1.31′	± 0.04′ = 3.5 %
48	477	+ 11.1	+ 1.75′	± 0.05′ = 3 „
43	539	+ 8.1	+ 1.28′	± 0.04′ = 3 „
40	574	+ 2.6	+ 0.41′	± 0.02′ = 6 „
52	599	— 2.3	— 0.36′	± 0.03′ = 8 „
45	637	— 6.0	— 0.95′	± 0.01′ = 1 „
51	688	— 4.9	— 0.78′	± 0.03′ = 4 „

greater than + 1,75′, but they still admitted of exact measurement. The above-mentioned change of sign is analogous to that found by INGERSOLL in the infra-red and presents a characteristic and theoretically important phenomenon.

The relation between the rotation and the field was also investigated, and the results are shewn in Table 2 and Fig. 3. For low values of the field the two are proportional to each other, the rotation

TABLE 2.

ε = funct ($\mathfrak{H}$) Cupriferrite λ = 477 $\mu\mu$

N	$\mathfrak{H}$ (kgs)	Δ (mm)	ε (Minutes)	$\delta\varepsilon$
40	0.93	+ 5.3	+ 0.85′	± 0.02′ = 2 %
22	2.25	+ 8.4	+ 1.34′	± 0.04′ = 3 „
59	4.47	+ 9.7	+ 1.56′	± 0.03′ = 2 „
31	7.19	+ 10.2	+ 1.63′	± 0.03′ = 2 „
20	9.32	+ 10.4	+ 1.66′	± 0.03′ = 2 „
48	10.15	+ 11.1	+ 1.75′	± 0.05′ = 3 „

afterwards assuming a maximum value, which remained nearly constant for further increase of the field. Considering the form of the curve ε = funct ($\mathfrak{H}$) and accepting the results previously found

by DU BOIS in the case of iron, nickel, and cobalt, we may assert the proportionality between ε and $\mathfrak{J}$ with great probability. Bearing in mind this fact we are able to determine from purely magneto-optic measurements the order of magnitude or at least an inferior limit of maximum magnetization. As DU BOIS [1]) has shewn in the case of an unlimited homogeneous plane disc, the magnetization of which is uniform and normal, the abscissa of the point of intersection of the straight line $\varepsilon = K\mathfrak{J} = K\mathfrak{H}/4\pi$ and of the asymptote $\varepsilon = const.$ has the value $4\pi\mathfrak{J}_m$.

Accordingly $\mathfrak{J}_m \geqq 140$ c.g.s. in the case of cupriferrite. The small inclination of the upper part of the curve in Fig. 3 may be explained by the fact that for irregularly formed specimens the real conditions do not correspond to those in the ideal case mentioned above. However this inevitable difference can only produce a decrease in the apparent value of $\mathfrak{J}_m$ so that an inferior limiting value is really determined; small fissures, cavities, and impurities in the reflecting surface are particularly capable of exerting such an influence.

2. *Magnetite.* The dispersion of the KERR-Effect is shown in Table 3 and Fig. 4 (continuous line). If we compare this curve with the

TABLE 3.

ε = funct (λ) Magnetite $\mathfrak{H} = 11.56$ Kgs

N	λ ($\mu\mu$)	$\angle$ (mm)	ε (Minutes)	$\partial\varepsilon$
30	436	— 24.0	— 3.81′	$\pm$ 0.03′ = 0.9%
25	442	— 19.9	— 3.15′	$\pm$ 0.05′ = 1.5 „
15	453	— 9.6	— 1.52′	$\pm$ 0.03′ = 2 „
30	464	0	0	—
40	477	+ 6.7	+ 1.06′	$\pm$ 0.03′ = 2 „
26	510	+ 19.4	+ 3.07′	$\pm$ 0.02′ = 0.6 „
25	539	+ 24.3	+ 3.84′	$\pm$ 0.02′ = 0.5 „
30	574	+ 28.2	+ 4.45′	$\pm$ 0.02′ = 0.4 „
30	599	+ 24.9	+ 3.94′	$\pm$ 0.02′ = 0.5 „
31	637	+ 21.0	+ 3.32′	$\pm$ 0.04′ = 1 „
30	688	+ 16.0	+ 2.50′	$\pm$ 0.07′ = 3 „

[1]) H. DU BOIS, Wied. Ann. **31** p. 965, 1887; Phil. Mag. (5) **29** p. 301, 1890.

previous one given by DU BOIS (dotted line), which he obtained with the same specimen (a holoëdric regular crystal, possessing a natural octahedral surface) we see, that with the exception of a displacement throughout the whole range of wave-lengths amounting to about 10 to 30 $\mu\mu$ — which is explained by the fact that 20 years ago only an imperfect method of spectral decomposition was available — the curves are in agreement in the region between 486 and 671 $\mu\mu$. The rotation attains a maximum value of 4.45′ in the yellow and decreases rapidly with decreasing wave-length. DU BOIS [1]), who was unable to proceed further than the blue on account of insufficient intensity of light, observed that the rotation probably vanished in the blue; he also considered that a change of sign possibly might occur in the ultraviolet. I have located this zero-point in the visible part of the violet at 464 $\mu\mu$. For smaller wave-lengths the rotation has rather a large *negative* value, which seems to approach a minimum. Unfortunately it was impossible to carry the investigation beyond 436 $\mu\mu$ since the light at that point becomes too feeble.

At all events, the existing observations establish satisfactorily the fact that the dispersion-curve obtained with natural crystalline magnetite ($FeO \,.\, F_2O_3$) is of the same type as that obtained above with cupriferrite. Without entering into theoretical considerations it may be seen at once that in both cases the curve passes through a maximum, goes through zero and probably also through a minimum. Experiments are being carried out to see whether the course of these curves depends on the optical constants of the substances investigated, viz. their ordinary absorption- and dispersion-curves.

In the same way as in the case of cupriferrite the relation between the rotation and the field was also investigated. The results are shown in Table 4 and Fig. 5. They give $\mathfrak{J}_m \geqq 358$ C. G. S., which agrees with that obtained by DU BOIS [2]) (350).

The magnetic properties of magnetite crystals have been recently investigated by QUITTNER [3]), adopting WEISS' methods. From his measurements it follows that the component magnetization parallel to the field, which in this case is alone of interest, reaches a saturation value of about 475 C. G. S.; this subject and the cause of the discrepancy ought to be investigated in greater detail. One remark, however, may be made at once. In many cases the natural magnetite slightly departs from the simple structural formula ($Fe\,0, Fe_2\,0_3$);

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[1]) H. DU BOIS l. c. p. 38.

[2]) H. DU BOIS. Phil. Mag. (5) **29**, p. 301, 1890.

[3]) P. WEISS, Journ. de Phys. (3) **5** p. 435, 1896 and (4) **9**, p. 373, 1910. V. QUITTNER, Dissertation, Zürich, 1908.

TABLE 4.

ε = funct ($\mathfrak{H}$) Magnetite $\lambda = 574\ \mu\mu$

N	$\mathfrak{H}$ (Kgs)	Δ (mm)	ε (Minutes)	$\delta\varepsilon$
15	2.10	+ 12.9	+ 2.07′	$\pm$ 0.04′ = 2 %
15	3.40	+ 21.3	+ 3.37′	$\pm$ 0.05′ = 1.5 „
15	5.87	+ 28.7	+ 4.54′	$\pm$ 0.07′ = 1.5 „
15	8.87	+ 28.0	+ 4.43′	$\pm$ 0.05′ = 1 „
15	10.82	+ 28.9	+ 4.57′	$\pm$ 0.06′ = 1 „
30	11.56	+ 28.2	+ 4.45′	$\pm$ 0.02′ = 0.4 „

also QUITTNER has established the great diversity of samples by measuring their variable densities. It is difficult to foretell the influence of all this on the magneto-optic properties.

3. *Other ferromagnetic compounds.* The distinct analogy in the dispersion for substances of similar chemical structure as e.g. cupri- and ferroferrite in contradistinction to iron, nickel, and cobalt suggests whether the properties of other ferromagnetic ferrites and oxides are not similar. The investigation of *calciumferrite* was in this respect of importance. This substance is very feebly magnetic and brittle. A small piece was surrounded by the easily fusible WOOD alloy and then thoroughly polished. No KERR-Effect however was observed although the mirror was sufficiently good. The effect, if it exists, must be smaller than 0,35′. A similar result was obtained with *ilmenite* [1]). The light was reflected from the base of the crystal, as well as from a plane parallel to the principal axis, but in no case could a rotation be detected. ($< 0{,}3'$). It was also impossible to detect any rotation with *chromic oxide* Cr_2O_3, which without doubt is ferromagnetic. The following alloys were tested:

4. *Nickel-iron* with 36% nickel, so called "Invar", known to possess a very small coefficient of expansion, is strongly magnetic and distinctly shows the KERR-Effect. The rotation is exclusively *negative* in the region of the spectrum investigated, and there is only a slight variation with wave-length. (Table 5, Fig. 6). The dispersion-curve lies considerably below the zero-line; with increasing wave-length

[1]) See B. BAVINK, Magn. Influenz in Krystallen, Göttinger Dissertation 1904.

TABLE 5.

ε = funct (λ) "Invar" $\mathfrak{H}$ = 13.30 Kgs.

N	λ ($\mu\mu$)	Δ (mm)	ε (Minutes)	$\delta\varepsilon$
15	436	— 74.4	— 11.78′	$\pm$ 0.05′ = 0.4%
15	477	— 78.8	— 12.48′	$\pm$ 0.06′ = 0.5 „
15	539	— 83.5	— 13.22′	$\pm$ 0.06′ = 0.4 „
20	574	— 86 3	— 13.66′	$\pm$ 0.03′ = 0.2 „
15	599	— 86.8	— 13.74′	$\pm$ 0.05′ = 0 3 „
15	637	— 86.7	— 13.72′	$\pm$ 0.07′ = 0.5 „
15	688	— 86.2	— 13.54′	$\pm$ 0.06′ = 0.4 „

it proceeds slowly downwards, passes through a flat numerical maximum in the orange, after which the rotation decreases very slowly. The relation between rotation and magnetization, as in the cases above, exhibits distinct proportionality and we have $\mathfrak{H}_m \geq 530$ (Table 6, Fig. 7).

TABLE 6.

ε = funct ($\mathfrak{H}$) "Invar" λ = 574 $\mu\mu$

N	$\mathfrak{H}$(kgs)	Δ (mm)	ε (Minutes)	$\delta\varepsilon$
31	0.54	— 6.5	— 1.02′	$\pm$ 0.02′ = 2 %
15	1.80	—23.2	— 3.67′	$\pm$ 0.02′ = 0.5 „
15	3.20	—39.0	— 6.17′	$\pm$ 0.03′ = 0.5 „
15	6.32	—69.7	—11.03′	$\pm$ 0.05′ = 0.4 „
15	10.37	—84.5	—13.36′	$\pm$ 0.03′ = 0.2 „
15	12.60	—86.6	—13.71′	$\pm$ 0.02′ = 0.1 „
20	13.30	—86.3	—13.66′	$\pm$ 0.03′ = 0.2 „
15	14.51	—86.2	—13.65′	$\pm$ 0.03′ = 0.2 „

It would be interesting to study the magneto-optic behaviour of the nearly non-magnetic nickel-iron alloy, which contains 25 percent nickel.

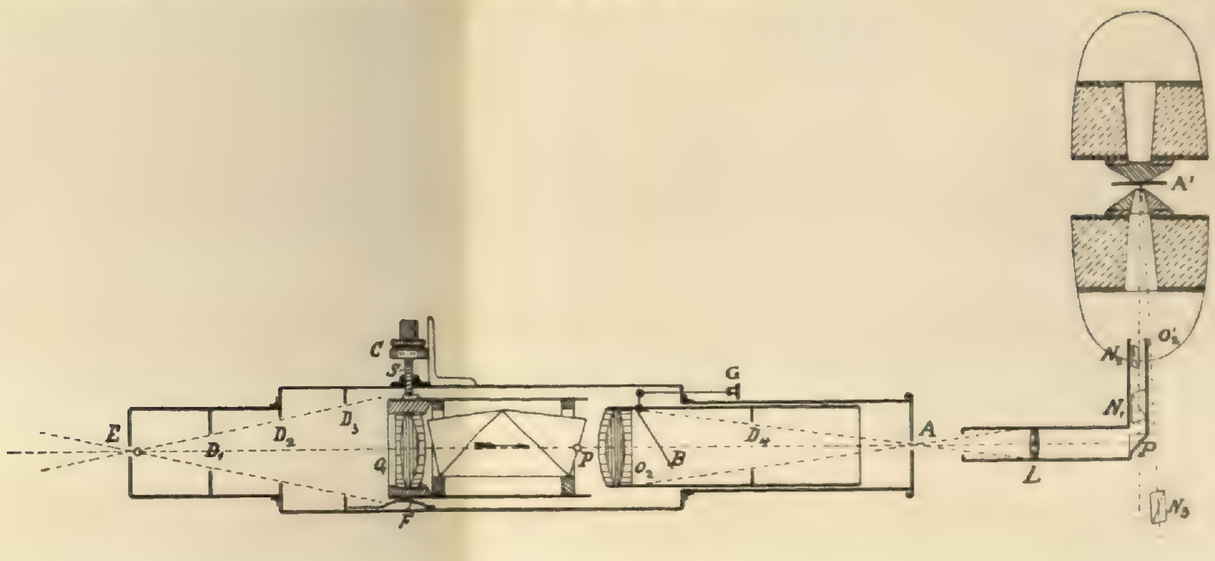

Fig. 1.

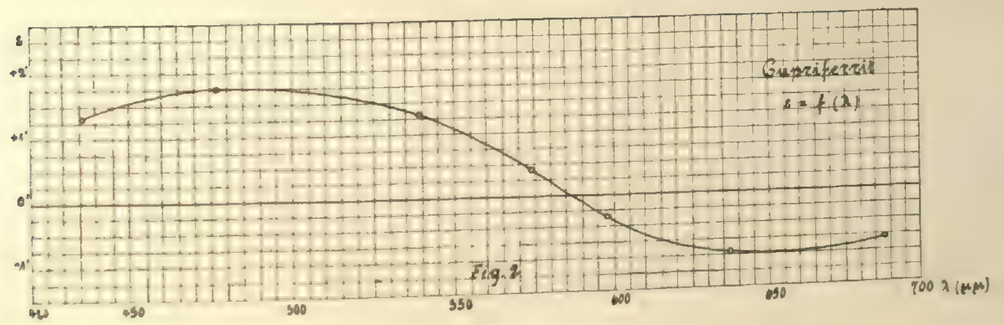

Fig. 2

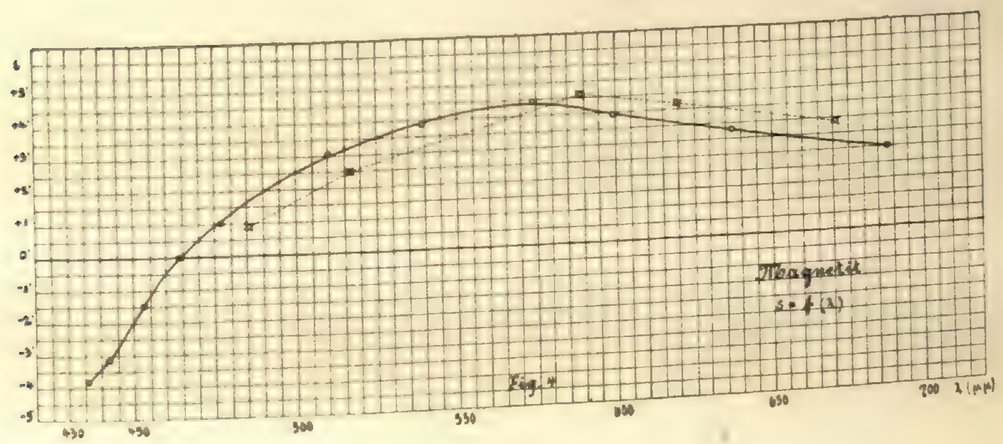

Fig. 4.

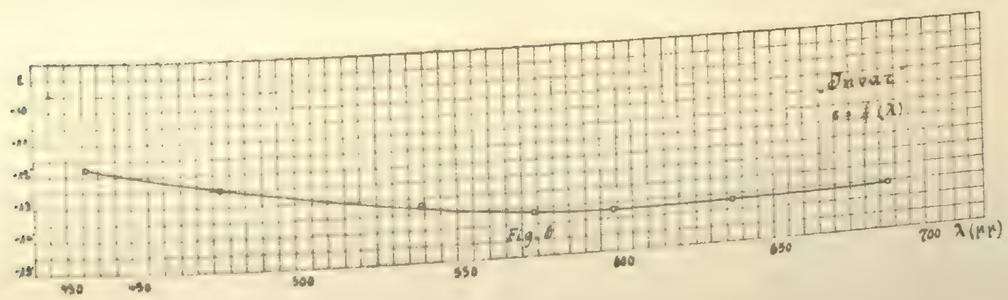

Fig. 6.

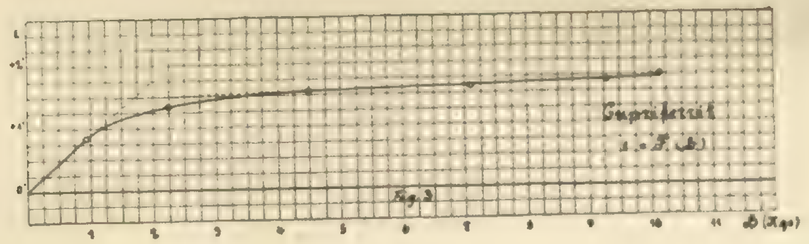

Fig. 3.

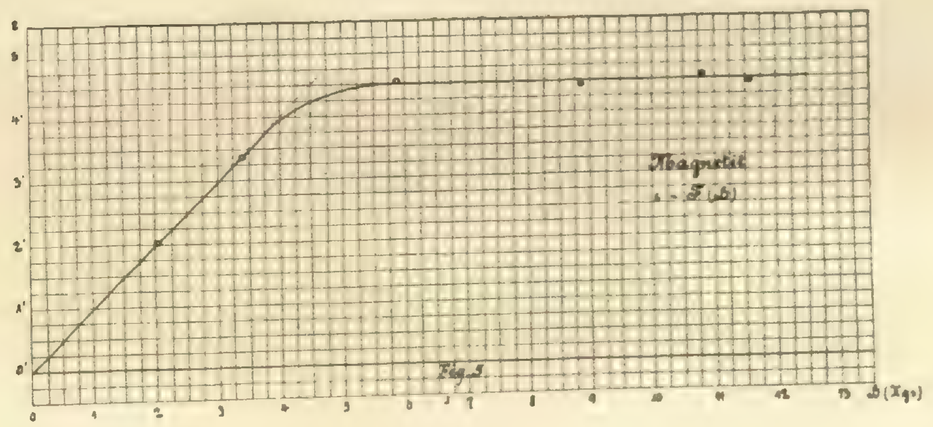

Fig. 5.

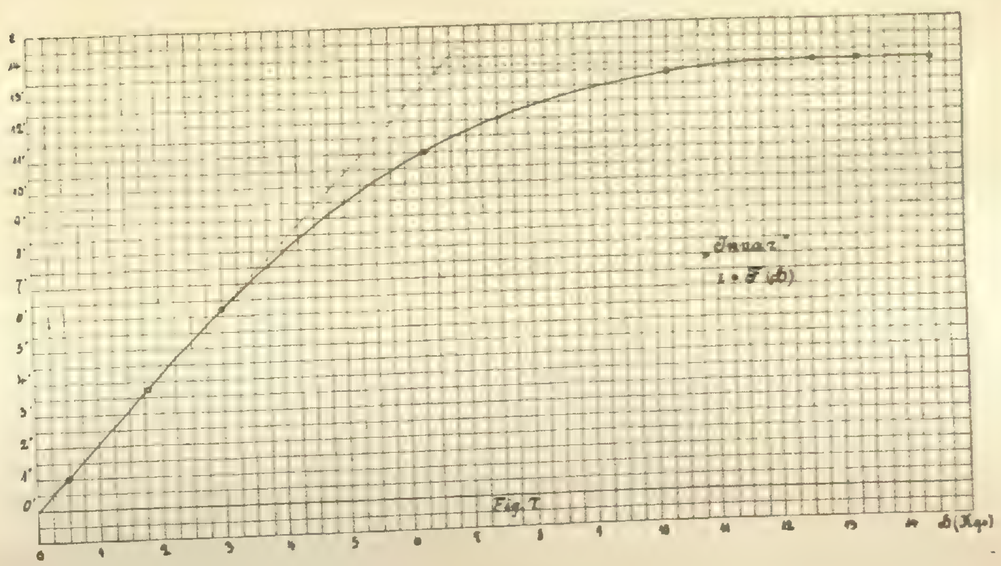

Fig. 7.

5. *The* HEUSLER *alloy*, supposed to contain 61% Cu, 26% Mn and 13% Al is rather strongly magnetisable. Different portions of two well-polished mirrors were carefuly examined in various parts of the spectrum but proved to be magneto-optically ineffective. It is of course possible that the KERR-Effect might be less than 0,3′ in this case. Quite recently there appeared a communication by TOKMATSCHEW recording similar experiments on the HEUSLER alloy No. 32 (58,9 Cu, 26,5 Mn, 14,6 Al). From theoretical considerations the author arrives at the conclusion of the probability of an effect capable of measurement occurring in the neighbourhood of 450 $\mu\mu$. I have carried out a series of readings at this wave-length but no rotation could be observed. INGERSOLL also failed to notice any measurable effect either in the visible spectrum or in the infra-red.

The discussion of the theoretical signification of the above partially positive and partially negative results I reserve for a future occasion; further experiments are in preparation, and the determination of the purely optical properties of the investigated substances is already in progress.

ERRATA.

In the Proceedings of the Meetings of Jan. and Febr. 1910:

p. 672 Table III for 5050 read 8050.
p. 675 Table VII for 102.58 read 102.85.
p. 676 Table VIII for 71.75 read 71.95.

(May 26, 1910).

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